

PATHWAYS AND ESTIMATED CONSEQUENCES OF RADIONUCLIDE RELEASES FROM A NUCLEAR POWER PLANT

A study related to
Barsebäck nuclear power plant

Mats Nilsson



Lund 1981



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A study related to Barsebäck Nuclear Power Plant

av

Mats Nilsson
Fil. kand., Mlm

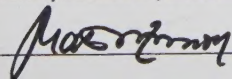
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Title and subtitle PATHWAYS AND ESTIMATED CONSEQUENSES OF RADIONUCLIDE RELEASES FROM A NUCLEAR POWER PLANT. A study related to Barsebäck Nuclear Power Plant.			
Abstract The radioactive corrosion products ^{60}Co, ^{58}Co, ^{57}Co, ^{54}Mn, ^{65}Zn, ^{51}Cr and ^{110}Ag which are released from Barsebäck Nuclear Power Plant (Sweden) into the marine environment have been studied. The brown seaweed <u>Fucus vesiculosus</u> has been found to be an excellent bioindicator for these radionuclides. The distribution of activation products along the west coast of Sweden has been studied and can very well be described with a power function. Uptake and retention of the activation products in <u>Fucus</u> have been studied and results are reported. The activity concentration in <u>Fucus</u> well reflects the discharge rate from the power plant. Other bioindicators have been studied; mainly the crustaceans <u>Idothea</u> and <u>Gammarus</u>. A simulation of ^{131}I-release following a fictitious BWR 1 accident is described as well as a model for estimation of the individual and collective dose equivalents arising from intake of contaminated milk.			
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PATHWAYS AND ESTIMATED CONSEQUENCES OF RADIONUCLIDE RELEASES FROM A NUCLEAR POWER PLANT

**A study related to
Barsebäck nuclear power plant**

Mats Nilsson



Lund 1981

PATHWAYS AND ESTIMATED CONCENTRATIONS
OF RADIOACTIVE RELEASES FROM
A NUCLEAR POWER PLANT

A study carried out
Dutchess County Nuclear Power Plant

State of New York



June 1981

To the memory of Curt Pettersson

This thesis is based on the following papers, which in the text will be referred to by their Roman numerals.

- I. Mattsson,S., Finck,R. and Nilsson,M.:
Distribution of activation products from Barsebäck nuclear power plant (Sweden) in the marine environment. Temporal and spatial variations as established by seaweed. Env. Poll. (Series B) 1, 1980, pp 105-115
- II. Nilsson,M., Dahlgaard,H., Edgren,M., Holm,E., Mattsson,S. and Notter,M.:
Radionuclides in Fucus from inter-Scandinavian Waters. In Proc. of IAEA Symposium on the Impacts of Radionuclide Releases into the Marine Environment, 1980.(In press)
- III. Nilsson,M., Mattsson,S. and Holm,E.:
Radioecological studies of activation products released from a nuclear power plant into the marine environment. Report CODEN:LUNDFD6/(NFRA-3025)/1-19/(1981).
- IV. Nilsson,M.:
Mathematical models for simulation of processes of radioecological and biomedical transport of radionuclides. Part III: Application on ^{131}I in the food-chain grass-cow-milk-man following a BWR 1 accident. To be published as a report from the Research Institute of National Defence, Umeå, Sweden.

Reports from papers I, II and III were given at:

Third NEA Seminar on Marine Radioecology, Tokyo, Japan
1-5 October, 1979.

5th International Congress of IRPA. Jerusalem, Israel
10-14 March, 1980

International Symposium on the Impacts of Radionuclide Releases to the Marine Environment. IAEA, Vienna, 6-10 October, 1980.

Second Nordic Seminar on Radioecology, Helsingør, Denmark
8-10 May, 1979.

<u>CONTENTS</u>	page
Introduction	1
The activation products - sources and production	3
Detection of the radionuclides and mapping of their distribution using bioindicators	8
Results and discussion	9
Simulation of ^{131}I -releases following a fictitious BWR 1 accident	14
Results and discussion	15
Acknowledgements	17
References	18

INTRODUCTION

The production of energy in different forms and by different means is always connected with some kind of hazard. It may result in negative effects on the sensitive ecosystems in the environment, as well as health effects on individuals and populations.

Discussions concerning these hazards have especially in the late 1970s almost entirely been focussed upon the production of energy using nuclear power.

On January 1, 1979, the number of operating nuclear power plants for commercial purposes throughout the world was 224 (IAEA, 1980). The estimated share of total electricity from these reactors was 7.8%. To this number (224) should be added an unknown (but great) number of reactors for research and military purposes.

The frequently used words "nuclear safety" has above all been connected with radiological matters. What is then the meaning of the word "safety", especially in connection with releases of radionuclides?

All handling of radioactive materials in the nuclear fuel cycle, from the uranium ore to the contents of the waste canisters, will give rise to releases of radionuclides to the environment. Sometime in the future, radionuclides emanating from the final deposition of the nuclear waste will again be introduced into the biosphere.

From the nuclear power companies' point of view, the meaning must involve to by all means ensure that any kind of accident will be highly improbable. The meaning also includes a responsibility to minimize the radioactive discharges to the environment. On the other hand, nuclear safety also demands that the supervising authorities in a proper and objective manner can control that these commitments are fulfilled. The practical analytical part of the control programmes should therefore be carried out by official authorities which are not connected to the nuclear power companies.

A third topic included in "nuclear safety" concerns the need for scientific data regarding radioactive elements, their behaviour, levels and pathways in the biosphere as well as effects on living organisms and populations. The maintenance of the knowledge in these sciences, known as radioecology

and/or environmental radiology, is of the utmost importance in a society dependent to such great extent on nuclear energy.

The need for knowledge regarding radionuclides and their behaviour in the environment is evident, and a number of people have throughout the world during about two decades been engaged in such research. There is, however, still a great need for reliable data on distribution patterns and transfer of radionuclides. It is for example extremely important to consider whether data from one site is applicable at another site, perhaps in another part of the world. It is clear that the properties of the radionuclides differ from source to source as well as from site to site. The new analytical techniques available have made it possible to detect still lower activity values as well as to extend the studies to other radionuclides of which, due to practical detection problems, up to now little has been known.

The purpose of this work was to study activation products released from the Barsebäck nuclear power plant in Southern Sweden. The results from this research is reported in papers I, II, and III. A study concerning collective and individual doses arising from the intake of ^{131}I via milk after a fictitious BWR 1 accident in the same power plant is found in paper IV.

The Barsebäck power plant is located on the Swedish west coast at the Öresund sound 20 km east of Copenhagen. It consists of two boiling water reactors, each of 1700 MW (thermal) effect. The first reactor became operational in April 1975 and the second in March 1977.

Releases of radionuclides of radiological importance from these reactors are made mainly to the marine environment. At the present degree of burnup the releases mainly consists of activation (corrosion) products. A continued operation of the plant will probably result in damage on the fuel encapsulation. This is quite normal, but will give rise to releases of fission products into the marine environment. These will probably mainly consist of iodine, cesium and ruthenium isotopes.

If an accident occurs, the amount of activity released will drastically be increased. The activity concentration will on the terrestrial side be higher and this side will also be subject to the most serious acute effects. The total amount of activity that is discharged to the marine environment will probably be considerably higher. The presence of this great amount of radioactive material in the water and sediments of the Öresund which will follow from such an accident will lead to an increased intake of radionuclides from fish consumption and aerosol spreading. This intake can give rise to considerable collective doses due to the large population exposed as well as considerable dose commitments due to resuspension of radionuclides from the sediments, which will require a long integration time in the dose calculations.

It is therefore of great interest and importance to study the spreading pattern of radionuclides from this power plant. For this purpose, the controlled, small releases of activation products has been used. The knowledge following such research can then be used in order to make forecasts regarding the activity distributions in case of a major release from the power plant.

THE ACTIVATION PRODUCTS - SOURCES AND PRODUCTION.

The activation products studied are all produced, mainly on the surfaces of the fuel elements, by neutron induced nuclear reactions. Some photonuclear reactions could occur in the main steam pipes due to the high energy photons from ^{16}N but the contribution is negligible.

The "target" atoms in these reactions are produced by corrosion in the reactor vessel, steam pipes, turbine, condenser, feed-water pipes etc. The corrosion rate is high, due to the high temperatures and pressures in these parts. In order to prevent corrosion, the reactor water is totally demineralized, and the oxygen content is kept as low as possible.

All activation products studied are isotopes of the following elements

Iron (Fe) (^{54}Fe , 5.82%, ^{56}Fe , 91.66%, ^{57}Fe , 2.19%, ^{58}Fe , 0.33%)

Cobalt (Co) (^{59}Co , 100%)

Manganese (Mn) (^{55}Mn , 100%)

Zinc (Zn) (^{64}Zn , 48.89%, ^{66}Zn , 27.81%, ^{67}Zn , 4.11%, ^{68}Zn , 18.57%, ^{70}Zn , 0.62%)

Chromium (Cr) (^{50}Cr , 4.31%, ^{52}Cr , 83.76%, ^{53}Cr , 9.55%, ^{54}Cr , 2.38%)

Silver (Ag) (^{107}Ag , 51.82%, ^{109}Ag , 48.18%)

The stable isotopes of these elements can be found in almost every part (metallic) in the reactor construction, but the main sources for corrosion and later activation in the Barsebäck reactors are for (Knutsson, Bliselius, Ahlmann and Nilsson, 1980):

- 1) Iron: the reactor vessel, steam pipes, turbine and feed-water pipes. The iron content of the reactor vessel (stainless) is about 60%. The rest of the components are not stainless and contain >98% iron.
- 2) Cobalt: the main sources for cobalt are the cones and seats of the valves in the primary coolant and feed-water pipes. These components are made of stellite, an alloy consisting of more than 55% cobalt. The welding material for Co-alloys also contain a large fraction of cobalt. The high corrosion rate at about 160° for the stainless steel in the high pressure feed-water heaters also contribute to the cobalt available for activation. It is therefore important to have a low cobalt content in these, which is lower than 0.05%.
- 3) Manganese: The main source is the reactor vessel and the steam pipes, which contain 2.0% and 0.45% manganese, respectively.
- 4) Zinc is only present in one structure in the system, namely the condenser tubes. In each condenser there are 32000 tubes made of aluminium brass (Al + Zn + Cu). Because of many tube failures these have gradually been replaced by titanium tubes. (This was made in Barsebäck 1 in 1980).
- 5) Chromium: all stainless steel parts consist of up to 19% chromium. The stellite components contain about 30% chromium. The parts used to hold the fuel-rods at correct position are made of Inconel, an alloy consisting of about 17% chromium.
- 6) Silver: This metal is used only as sealing material between valves and pipes.

One element more should be mentioned: nickel (Ni). The main source for this is the stainless steel (14% Ni) and the Inconel parts (70% Ni).

These corrosion products (stable and radioactive) are present mainly in these systems.

- 1) The primary coolant
- 2) The condensate
- 3) The feed-water
- 4) The water in the spent fuel basins.

These systems are being constantly purified in ion-exchangers. The effect of such ion-exchangers can be seen in the following tables (primary coolant = prior to the condenser, feed-water = after the condensate purification) (Elkert, 1979).

Table 1

Corrosion products in primary coolant ($\mu\text{g/kg}$)

	Fe	Cr	Ni	Zn	Total
Barsebäck 1	1.4	2.3	0.37	0.33	5.2
Barsebäck 2	1.3	4.7	0.48	0.30	7.9

Table 2

Corrosion products in feed-water ($\mu\text{g/kg}$)

	Fe	Cr	Ni	Zn	Total
Barsebäck 1	0.6	0.06	0.16	0.04	1.0
Barsebäck 2	0.5	0.07	0.15	0.04	0.9

The specific radionuclides studied will now be presented.

- A. ^{59}Fe . This nuclide is produced in the reactions $^{59}\text{Co} (n,p) ^{59}\text{Fe}$ and $^{58}\text{Fe} (n,\gamma) ^{59}\text{Fe}$. It has a half life of 44.6 d and is a β^- and γ emitter. The activity produced is low and the discharges are negligible.
- B. ^{60}Co . This radionuclide is produced in the reaction $^{59}\text{Co} (n,\gamma) ^{60}\text{Co}$. It has a half life of 5.27 a and is also a β^- and γ emitter. The activity produced is one of the highest of the nuclides studied and the ^{60}Co -activity dominates the discharges into the sea (III).

- C. ^{58}Co is produced in the nuclear reactions $^{58}\text{Ni} (n,p) ^{58}\text{Co}$ and $^{59}\text{Co} (n,2n) ^{58}\text{Co}$. It decays through β^+ (85%) and EC (15%) and is also a γ emitter. Its half life is 70.8 d. The ^{58}Co -activity is in the reactor water the second highest produced and the activity in the discharges is also one of the highest.
- D. ^{54}Mn is the result of the reaction $^{54}\text{Fe} (n,p) ^{54}\text{Mn}$. It decays with EC and is a γ emitter with a half life of 312 d. The activity in the reactor water as well as in the liquid discharges is one of the lowest of the activation products.
- E. ^{65}Zn is produced in the reaction $^{64}\text{Zn} (n,\gamma) ^{65}\text{Zn}$. It decays with EC (98.5%) and β^+ (1.5%) and is also a γ emitter. Its half life is 244 d. The activity produced in the reactor depends on the material in the condenser tubes. In Barsebäck its activity was until summer 1980 2-3 times higher than ^{54}Mn in the reactor water and discharges.
- F. ^{51}Cr is the radionuclide resulting from the reaction $^{50}\text{Cr} (n,\gamma) ^{51}\text{Cr}$. Its half life is 27.7 d and the decay is EC with following γ -emission. The ^{51}Cr -activity in the reactor-water is the highest of the nuclides studied but, due to its short half life, is in the liquid discharges comparatively low.
- G. $^{110}\text{Ag}^m$ is produced from $^{109}\text{Ag} (n,\gamma) ^{110}\text{Ag}^m$. It decays with β^- (98.7%) directly to excited states of ^{110}Cd and with IT (1.3%) to ^{110}Ag , which decays with β^- (100%) to other excited states of ^{110}Cd . γ -emission follows both decays, naturally. The half life of $^{110}\text{Ag}^m$ is 253 d, and the activity produced and discharged is one of the lowest of the activation products.

A relative comparison between the activity levels for the activation products studied is given in table 3. All values (typical) are normalized to ^{60}Co .

Table 3

	⁵⁹ Fe	⁶⁰ Co	⁵⁸ Co	⁵⁴ Mn	⁶⁵ Zn	⁵¹ Cr	¹¹⁰ Ag ^m	
Reactor water	0.2	1	6	0.5	1	130	1	
Discharges	1976	-	1	1.95	0.15	0.22	1.29	0.11
	1977	-	1	0.47	0.08	0.25	0.49	0.07
	1978	0.01	1	0.63	0.07	0.19	0.61	0.07
	1979	0.01	1	0.34	0.09	0.23	0.87	0.04
	1980	0.002	1	0.2	0.04	0.16	0.11	0.008

For all nuclides, the ratio to ^{60}Co differs between reactor water and discharge in table 3. To understand this, it is necessary to discuss the origin of the liquid discharges.

The radioactive liquid waste in the power plant can roughly be divided into the following categories:

- I) Clean water with fluctuating activity levels as system drainage, fuel-basin water etc.
- II) Contaminated water with low activity as floor-drainage, water from laundries and other cleaning-water.
- III) Chemical waste water with usually high activity as decontamination fluids, water from laboratories etc.
- IV) Waste in form of sludge with high activity as spent filter - and ion - exchange masses.

Water of category I is purified in ion-exchangers and filters. After testing, the water is normally returned to the system. Water of categories II and III is normally filtered or evaporated prior to storage in the discharge tanks. Waste water of category IV is solidified in bitumen or concrete canisters.

The liquid discharges is therefore consisting only of category II and III waste, which explains the pulsed pattern of the discharges, which are concentrated to the revision (or overhaul) periods, normally from June - October.

The mean residence time for the activation products in the waste handling until discharge is, averaged over the year in the order of 1-3 months (Bliselius, 1980). This, together with different efficiencies in the ion-exchangers can explain the differences between reactor water and discharges in table 3 for most nuclides. For ^{58}Co , however, this at first seems puzzling. The time delay can not explain the reduction in the ratio to ^{60}Co by a factor 10. However, ^{58}Co is mainly produced in the reaction $^{58}\text{Ni}(n,p)^{58}\text{Co}$, which could result in another chemical state for ^{58}Co , resulting in a different chemical behaviour in the ion-exchangers and filters (III).

From table 3 it is also evident that the amount of short-lived radionuclides in the discharges have substantially been reduced since 1979. This is the result of an improved waste handling system, resulting in better filtration and reduced volumes in the discharge tanks, thus giving lower values and longer mean residence times in the tanks (Bliselius, 1980).

DETECTION OF THE RADIONUCLIDES AND MAPPING OF THEIR DISTRIBUTION USING BIOINDICATORS.

The activation products discharged are being dispersed into enormous volumes of water. Because of the extremely low activity concentration of the activation products present in the sea-water, in the order of 10^{-3}Bq/kg , the use of it is impractical for detection of the nuclides. It is also impractical because of difficulties to obtain representative samples, as discussed in (II). Therefore, in order to detect the radionuclides and map their distribution in the environment, suitable bioindicators are needed. Furthermore, it is of great advantage if these objects are parts of food-chains finally leading to man, in order to estimate the radiological effects.

Such objects are fish and algae, Fish is from a radiological point of view the most interesting bioindicator, but since fish is, more or less, a non-stationary organism it is almost impossible to obtain samples under controlled, natural conditions.

Algae, however, are excellent bioindicators for these purposes. Due to their high concentration ability, about 10 000 (II), and the

fact that they are located at the same spot during their lifetime, makes them very suitable for such studies.

In this study, the brown seaweed Fucus vesiculosus has been the bio-indicator commonly used (I, II, III). Other bioindicators have also been studied, namely the seaweed Ascophyllum and Cladophora. The living animals included in this study are the crustaceans Idothea and Gammarus and the common mussel Mytilus Edulis.

RESULTS AND DISCUSSION

a) Distribution of activation products in the marine environment.

From studies using Fucus v. it is evident (I, II, III) that the activity concentration $C(z)$ at distance z , in water (and Fucus) north of the discharge pipe can be described with a power function:

$$C(z) = \alpha \cdot z^{-\beta} \quad (1)$$

where the constant α varies with the amount of activity that is discharged (III). The constant β has been found to be around 1.4 for all nuclides except $^{110}\text{Ag}^m$. The reason for this is discussed in article (III) and is probably due to the complex chemistry of silver. The variation of the constant β during four years, 1977-1980, is very slow, but a tendency that can be seen is that the value of β is slightly dependent on the activity discharged recently prior to sampling (III).

The uptake in Fucus per unit activity released has been found to be approximately the same for all nuclides except for silver and chromium, of which only 30 and 10% relative to the other nuclides, are taken up by Fucus v. These differences are also believed to depend on differences in chemical behaviour. For chromium, for instance, this could already be seen in the ion exchangers at the power plant.

The dynamics in uptake and retention in Fucus v. has been studied especially for the cobalt isotopes (I, II). The results give an effective half life for cobalt in Fucus of about 50-100 d (I, II). This can mainly be ascribed to growth dilution, but some elimination process evidently occurs, since the measured and calculated results agree well in the period of low growth-rate (autumn and winter). (II).

The uptake of activation products has been studied also in other algae (III). The picture is here different compared to Fucus v.. For instance, Cladophora shows a higher uptake of ^{59}Fe than Fucus while Ascophyllum has a lower uptake of the nuclides than Fucus (III). For the crustaceans, the ability to concentrate silver is exceptional (III). The commonly used Mytilus Edulis shows, regarding all activation products studied, lower activity concentration values than does Fucus v. The result is, considering the availability and sample preparation, that Fucus v. is an outstanding bioindicator for mapping the distant spreading of activation products.

The results from this research could be used in order to mathematically describe the transport of the radionuclides in Öresund. However, one very important compartment is the sediments. In this work, too little data (III) is available for the sediments in order to construct a mathematical model. Hopefully, more detailed sediment data will be available in the near future (Edgren, 1980).

b) Radiological effects of releases of activation products.

The radiological effects of the releases of activation products strongly depend on their presence in food-chains. Food-chains of interest are

- 1) Benthic microalgae - crustaceans - juvenile plaice
- 2) Phytoplankton - mussels - adult plaice
- 3) Macro algae - crustaceans - cod

which all finally lead to man.

The radionuclides of interest are ^{60}Co , ^{58}Co and ^{65}Zn .

As mentioned earlier, it is almost impossible to obtain representative results for the activity concentration in fish. According to data from Grimås (1979) and IAEA (1961) the activity concentration in fish is for cobalt roughly a factor of 20 lower than that for Fucus v. From these papers, it is also found that the concentration factors for ^{65}Zn in fish (meat) is about two to three times higher than for cobalt isotopes. Since the concentration values for ^{65}Zn in Fucus in these papers are found to be about a factor of 2 lower than found in this work (I, II), the conclusion must accordingly be, that

in the Öresund, the concentration factor for ^{65}Zn in fish (meat) can be about five times higher than for ^{60}Co and ^{58}Co . Since the activity concentration values in Fucus v. for ^{65}Zn is about 20% of the values for ^{60}Co , the daily (yearly) intake in activity units must be about the same for ^{65}Zn as for ^{60}Co .

Assuming, conservatively, that a person is consuming 1 kg per week of fish (meat) that always have been living about 0.5 km north of the discharge pipe in Barsebäck, this would give rise to yearly intakes of about 10 kBq/a for ^{60}Co , 2 kBq/a for ^{58}Co and 10 kBq/a for ^{65}Zn . This corresponds to 0.05% (^{60}Co), 0.03% (^{58}Co) and 0.1% (^{65}Zn) of the annual limits for intake by workers (ALI) as specified by the ICRP (ICRP, 1980). Using the information found in the ICRP publication that these radionuclides are approximately equally distributed within the body, these activity values would correspond to dose equivalent values of 5 μSv , 3.2 μSv and 10 μSv per year, respectively.

A simple and approximate method to calculate the dose equivalent values is to use the retention functions for these elements as given in the ICRP publication. These are:

For cobalt (time - unit: days)

$$R(t) = 0.5 \cdot e^{-\frac{\ln 2}{0.5}t} + 0.3 \cdot e^{-\frac{\ln 2}{6}t} + 0.1 \cdot e^{-\frac{\ln 2}{60}t} + 0.1 \cdot e^{-\frac{\ln 2}{800}t} \quad (2)$$

and for zinc

$$R(t) = 0.25 \cdot e^{-\frac{\ln 2}{12}t} + 0.75 \cdot e^{-\frac{\ln 2}{320}t} \quad (3)$$

For input of one activity unit per day, this will lead to the following differential equation system: (the effective removal constants are called K_i , for instance $K_1 = \frac{\ln 2}{0.5} + \lambda$ and the fraction of the activity that is excreted from compartment i is called f_i).

$$\frac{dA_i}{dt} = f_i - k_i A_i \quad (4)$$

to which the solutions are

$$A_i(t) = \frac{f_i}{k_i} (1 - e^{-k_i t}) \quad (5)$$

the equilibrium values are, of course

$$A_i(\infty) = \frac{f_i}{k_i} \quad (6)$$

As can be seen from (2) the 800 d - component for cobalt is of critical importance for a long lived nuclide as ^{60}Co . The equilibrium values will in this case be improbably high, as can be seen in table 4 below. An analysis excluding the 800 d - component has thus also been made. It should yet be mentioned that the results not are in agreement with data taken from standard man (ICRP, 1975) which give an equilibrium value of a factor four times the daily intake (stable cobalt). The equilibrium values for zinc are in good agreement with those found in "standard man" (177 times the daily intake of stable zinc).

Table 4

Total body content for a daily intake of one activity unit. The figures in parenthesis indicate percentage of equilibrium

Time (years)	^{60}Co	$^{60}\text{Co}^1)$	^{58}Co	^{65}Zn
0.5	26(28)	18(90)	15.6(93)	94(61)
1	40(43)	19(95)	16.7(100)	130(84)
2	60(64)	20(100)	16.7(100)	150(97)
5	85(90)	20(100)	16.7(100)	154(100)
10	94(100)	20(100)	16.7(100)	154(100)

1) without the 800 d-component

The 10 kBq/a (27 Bq/d) intake of ^{60}Co will result in an equilibrium value of

$$\left. \begin{array}{l} 27 \cdot 94 \approx 2500 \text{ Bq, or} \\ 27 \cdot 20 \approx 540 \text{ Bq, or} \end{array} \right\} \text{ICRP 30}$$

$$27 \cdot 4 \approx 100 \text{ Bq, (Standard man)}$$

The decay of ^{60}Co is β^- , with a mean particle energy of 100 keV. According to MIRD, the absorbed fraction of 1.25 MeV photons from a source uniformly distributed in the body is about 0.3. This will, per decay, add about 0.75 MeV to the energy deposited in the body. This will give yearly dose equivalents of 150 μSv , 34 μSv and 6.1 μSv , respectively, using the different equilibrium values discussed previously.

For ^{58}Co , which is ingested at a rate of 14 Bq/d, a similar analysis will result in dose equivalent values of 6 $\mu\text{Sv/a}$ and 1.4 $\mu\text{Sv/a}$, respectively.

For ^{65}Zn , the ingestion rate would, due to the higher concentration values in fish, be about 27 Bq/d, which will result in an equilibrium whole body content of 4200 Bq. The absorbed dose almost totally comes from the gamma rays following the decay, and with the appropriate absorbed fraction, 0.3, will result in a yearly dose equivalent of about 50 $\mu\text{Sv/a}$. It should be noted, however, that as for ^{60}Co , a long time is required until a "steady state" condition in the body exists (see table 4).

An assumption that perhaps better describes the real situation is that the activity levels in fish caught in the Öresund more likely are related to mean activity values. The activity and dose values would then reduce to (60 km up to the Kattegat).

$$\frac{\int_0^{60} C(z) dz}{\int_0^{60} dz} = \frac{\alpha}{60} \int_0^{60} z^{-1.4} dz \approx 0.2 \alpha \quad (7)$$

that is, 20% of those given above. It must be stressed that this analysis is very conservative and crude, since the activity distribution is known only at the coasts of the Öresund. The figures are, therefore, probably upper limits.

Another situation where irradiation of man could be important is from seaweed on shores. The dose rate in air one meter above ground for an uniformly distributed source on a surface is

$$\dot{D} = \frac{A}{2} \cdot \overline{hV} \cdot \left(\frac{\mu_{en}}{\rho} \right)_{\text{air}} \cdot \int_0^{\infty} \frac{r}{1+r^2} e^{-\mu \sqrt{1+r^2}} dr \quad (8)$$

where A is the area content (Bq/m²).

Assuming that the area content is 10 kg (w.w.)/m² of Fucus v. (2 kg d.w./m²), the maximum surface activity of ⁶⁰Co (the only radionuclide of interest) could be 6000 Bq/m² near the plant. Furthermore, assuming that only 1/20 of the area is covered with Fucus (opening angle 10° from the centre of a circle), this will give rise to an absorbed dose rate in air of

$$\dot{D} \approx 20 \mu\text{Sv/a}$$

which will result in low absorbed dose equivalents due to the limited time people are spending on the shores.

SIMULATION OF ¹³¹I-RELEASES FOLLOWING A FICTITIOUS BWR 1 ACCIDENT

In paper IV an attempt has been made to estimate the activity levels in milk and the following individual and collective dose equivalents arising from consumption of milk. The technique in this work sometimes utilizes crude approximations and assumptions. Much important data is at present not accurately known, for instance the geographical spreading of the activity arising from transport from and between the dairies. Another example is the variation of the age-distribution of the consumers between different parts of the area to which contaminated milk is delivered. The time-variation of activity concentration in the forage and pasture is another parameter where

approximations had to be made. This work should therefore be regarded to be applicable more in the radiation protection discipline than in the field of radioecology.

The situation described in paper IV (a BWR 1 accident) is highly improbable, and the fact that people should be allowed to consume the contaminated milk after such an accident is totally impossible. Yet the analysis has been made using this scenario, because of the simplicity in applying the input data. The analysis could very easily be applied to situations which are more probable, for instance lower activity releases, down to normal operating levels, or situations where the discharges are distributed differently in time. The technique is, however, basically the same.

RESULTS AND DISCUSSION

Due to the nature of the scenario used, which describes a highly improbable event, it is not possible to give actual results regarding radiological effects. The collective and individual doses given in paper IV should therefore rather be used as "starting values" for analysis of a more probable situation. All correction factors to be used, for instance those given in the paper, are correcting the result in direct proportion to their magnitude.

As an example, a less unlikely event is that 10% of the iodine inventory is released, and that the milk production in the dairies is stopped during, say, two months. All dose figures would then reduce to about 10^{-4} of those given as "starting values". The maximum collective dose equivalents would then reduce to about 10^4 - 10^5 man Sv (summer) and 10^{-1} - 10^0 man Sv (winter). If the contaminated milk is uniformly distributed to the consumers ($\approx 10^6$ persons) the average individual dose equivalents would be around 10^{-2} - 10^{-1} Sv (summer) and 10^{-7} - 10^{-6} Sv (winter).

The individual doses would reduce by the same factor only if the milk that is consumed is delivered from dairies.

There are, of course, other iodine isotopes which are released in connection with such an accident. The most important of these is ^{133}I , but due to its short half-life (21 h), the contribution from this radionuclide to all dose figures will be small.

In order to improve this analysis complementary data is required. At present, information giving the missing data mentioned above is being studied (Wickman, Finck, 1980)

Data concerning the atmospheric spreading can also be improved, because of the site-specific characteristics of the spreading-pattern. A tool which recently has been developed for this purpose is the extremely sensitive technique of analysis of sewage sludge for the detection of atmospherically transported radionuclides (Ingemansson et al., 1980). This method of analysis is at present used in the surroundings of three Swedish nuclear power plants and seems, indeed, to be very promising.

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DISTRIBUTION OF ACTIVATION PRODUCTS FROM BARSEBÄCK NUCLEAR POWER PLANT (SWEDEN) IN THE MARINE ENVIRONMENT. TEMPORAL AND SPATIAL VARIATIONS AS ESTABLISHED BY SEAWEED

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ABSTRACT

*The biophysical behaviour of the radioactive corrosion products ^{51}Cr , ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn and $^{110}\text{Ag}^m$ has been studied in the marine environment up to 150 km from a nuclear power station using the brown seaweeds *Fucus vesiculosus* and *F. serratus* as bioindicators.*

*The activity concentration in *Fucus* per unit activity released from the nuclear power station is about equal for ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn but lower by a factor of 3 for $^{110}\text{Ag}^m$ and by a factor of 10 for ^{51}Cr .*

*The biological half-time of radioactive cobalt in naturally growing *F. vesiculosus* was 60 ± 15 days during the summer. Approximately the same value was found for ^{54}Mn and ^{65}Zn .*

Our results show that 85–90% of the activity released from the nuclear power plant follows the dominating north-bound water stream and 10–15% is transported southwards.

For ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn the decrease in activity concentration $C(z)$ with distance, z (km) north of the power plant can be described by a power function, $C(z) = \alpha \cdot z^{-1.4}$, where α is a constant.

INTRODUCTION

The marine environment is the main recipient of radioactive substances released from the nuclear power industry. To be able to estimate the radiological consequences of such releases, it is of fundamental interest to know how the radionuclides released are transported in water and how they are transferred from water to different organisms in the sea and, finally, to man. Knowledge of the

transport routes and of transfer coefficients for inter-Scandinavian waters is not yet adequate enough to allow a proper estimation of the collective dose commitment to man. It is therefore of interest to use the strictly controlled releases from nuclear power stations already in operation to permit better estimates to be made of the radiological consequences of presumably higher future releases.

In this work we have used the seaweed *Fucus vesiculosus* as a biological indicator of the temporal and spatial variations in the concentration of activation products released from the Barsebäck nuclear power plant into the marine environment.

The first indication of a more distant spread of radioactive material from Barsebäck was gained in the summer of 1976 when ^{60}Co was detected in *Fucus serratus* collected at Särö (56° 76' N, 12° 63' E) on the west coast of Sweden, where we have been studying the long-term variation of the ^{137}Cs -activity concentration in the marine environment since 1967 (details to be published later).

CHARACTERISTICS OF THE BARSEBÄCK NUCLEAR POWER PLANT AS A SOURCE OF RADIONUCLIDES FOR RADIOECOLOGICAL STUDIES IN THE MARINE ENVIRONMENT

The Barsebäck nuclear power plant is located on the Öresund between Denmark and Sweden, 18 km north of Malmö (Fig. 1). It has two boiling water reactors, each with an electrical power output of 580 MW. The first reactor (B1) has been in operation since early 1975 and the second (B2) since March 1977. Information on the monthly release of different radionuclides into the Öresund is estimated by the plant laboratory at Barsebäck from monthly measurements on a mixed sample, composed of volume-proportional aliquots of the daily releases during a month (Sydkraft, 1975-78). In general, the discharge of radioactive material is slight during normal operation of the plant but is considerably higher during the weeks in summer when partial refuelling and service are carried out. This gives rise to a 'pulsed' release-rate pattern of radionuclides which, since it simplifies the evaluation of data, is very useful for studies of the uptake and retention of various radionuclides in marine ecosystems.

MATERIAL AND METHODS

Samples of the brown seaweed *Fucus vesiculosus* were collected along the shore at the localities indicated in Fig. 1. The depth of the water varied between 0.5 and 1 m. All seaweed collected was firmly rooted to the bottom on stones. After collection, the seaweed was sorted to remove species other than *F. vesiculosus*. After drying in air for 2-3 days and grinding, the samples were packed in 180- or 2000-ml cylindrical plastic containers and measured by 100, 60 or 46 cm³ Ge(Li)-detectors. Counting times varied from 5 to 100 h. The detection efficiencies of the Ge(Li)-spectrometers were carefully determined using samples containing known activities of ^{152}Eu , ^{22}Na



Fig. 1. Map of southwest Sweden and east Denmark. The two nuclear power stations (□) Barsebäck and Ringhals, as well as our present sampling stations for *Fucus*, etc. (●) are indicated.

and ^{57}Co . Corrections to the efficiency due to differences in density between different samples were made. After measurement, samples were dried at 105°C for 24 h to obtain their reference weights.

RESULTS AND DISCUSSION

*The time variation of the activation product concentration in *Fucus vesiculosus* and its relationship to releases from Barsebäck*

Figure 2 gives the activity concentration, on a dry weight basis, of ^{54}Mn ($T_{1/2, \text{phys}} = 312$ days), ^{58}Co (70.8 days), ^{60}Co (5.27 years) and ^{65}Zn (244 days) in *F.*

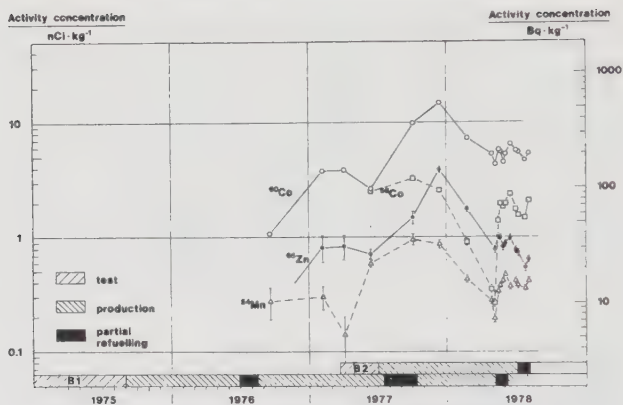


Fig. 2. Time variation of activation product concentration, on a dry weight basis, in *Fucus vesiculosus* at 2.9 km north-northwest of the Barsebäck discharge pipe. The timetable for testing, commercial production and refuelling of the two boiling water reactors. B1 and B2, is given on the abscissa.

vesiculosus collected between 1976 and 1978 at the shore at a point 2.9 km north of the Barsebäck discharge pipe. The periods during which testing, commercial energy production and refuelling of the two reactors, B1 and B2, took place are indicated in the abscissa. In addition to the activation products mentioned above, detectable concentrations of ^{51}Cr (27.7 days), ^{57}Co (272 days) and $^{110}\text{Ag}^m$ (252 days) were sometimes found.

The four activation products shown in Fig. 2, ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn , show similar time variation patterns in *F. vesiculosus* and the variations in activity concentrations seem to reflect the variation in discharge rate from Barsebäck, the latter having a pronounced maximum during periods of refuelling and service.

The two periods, August–October 1977 and May–July 1978, have been used to make a rough estimate of the relationship between the increase in activity concentration in *F. vesiculosus* and the activity of various radionuclides released from Barsebäck. The increase in activity concentration in *F. vesiculosus* per activity unit released is approximately the same for ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn . However, the $^{110}\text{Ag}^m$ activity concentration in *Fucus* is about a factor of 3 lower than expected in comparison with these four radionuclides. The concentration of ^{51}Cr in *Fucus* per activity unit released is only about 10% of that for ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn .

During the partial refuelling of B1 in the period May–June 1978, a more detailed

comparison between the output from Barsebäck and the activity concentration in *F. vesiculosus* was carried out for ^{58}Co . Information on the daily release of different radionuclides is unfortunately not available, only the 'total β -activity' of the individual (normally daily) releases being measured. To estimate the daily release of ^{58}Co , $a(t)$, we have assumed a constant ^{58}Co -activity/total β -activity ratio in the daily releases during a month.

Assuming that the concentration of ^{58}Co in the water surrounding the seaweed varies proportionally with the discharge rate, the time-dependent ^{58}Co -activity concentration, $C(t)$, in *F. vesiculosus* may be expressed as:

$$\frac{dC(t)}{dt} = ka(t) - (l + \lambda)C(t) \quad (1)$$

where k is a constant, describing the increase in activity concentration in *F. vesiculosus* 2.9 km north-northwest of Barsebäck after a release of unit activity (kg^{-1}), l is a biological elimination rate constant (day^{-1}) and λ the physical decay rate constant (day^{-1}).

Using different values of k and l , numerical integration shows that the best fit to the experimental points is given by $k = 2.1 \cdot 10^{-8} \text{ kg}^{-1}$ and $l = 1.2 \cdot 10^{-2} \text{ day}^{-1}$ (Fig. 3). This means that the biological half-life is about 60 days, with an estimated uncertainty of ± 15 days.

The radionuclide ^{57}Co , which has not as yet been detected in the liquid release from the plant, has been detected in *F. vesiculosus* at our sampling plots near Barsebäck at various times from February to August 1978. The ^{57}Co concentration varies with the distance from Barsebäck in the same way as ^{60}Co . This indicates that the ^{57}Co might also come from Barsebäck. The $^{57}\text{Co}/^{60}\text{Co}$ activity ratio was found to be 0.0024 ± 0.0002 . By means of this ratio and the known ^{60}Co release, we can estimate that ^{57}Co release during the first half of 1978 has been 1.2 mCi (44 MBq). The transfer of radioactive cobalt isotopes from water to *Fucus* will be the subject of further studies by comparative measurements of ^{60}Co , ^{58}Co —and possibly also ^{57}Co —from Barsebäck in water and *F. vesiculosus* (details to be published later).

The results of our studies so far indicate that the activation products ^{54}Mn , ^{58}Co , ^{60}Co and ^{65}Zn all have about the same biological half-life, 45–75 days, in *F. vesiculosus*. For the cobalt isotopes, this is in good agreement with the results of laboratory experiments on different brown, green and red algae (Nakahara *et al.*, 1975). For ^{65}Zn , however, the results of laboratory experiments show a higher retention of ^{65}Zn than that found here. Biological eliminations with a half-life of > 100 days (Gutknecht, 1965) and no significant elimination during 100 days (Young, 1975) have been reported from laboratory experiments on *F. vesiculosus* and *F. serratus*, respectively. Our results indicate a considerably higher elimination rate for ^{65}Zn in *F. vesiculosus* under actual natural conditions $T_{1/2, \text{biol.}} \approx 60$ days.

The radionuclides discussed above are all produced outside the fuel element

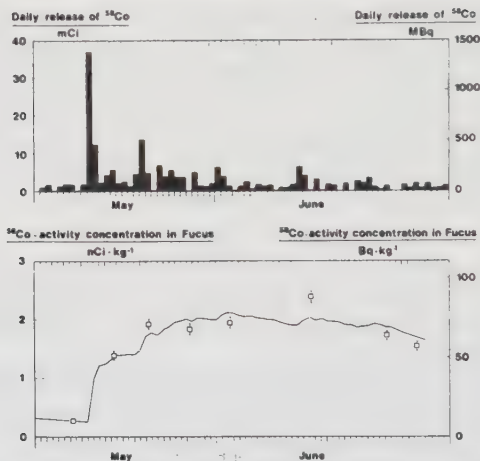


Fig. 3. Estimated daily release of ^{59}Co from Barsebäck in May-June 1978 and measured ^{59}Co -activity concentration ($\square$) in *Fucus vesiculosus* 2.9 km north-northwest of Barsebäck. The best fit of eqn (1) to the experimental data (solid line) was obtained for $k = 2.1 \cdot 10^{-9} \text{ kg}^{-1}$ and $l = 1.2 \cdot 10^{-2} \text{ day}^{-1}$ which means a biological half-life for cobalt in *Fucus vesiculosus* of 60 days.

cladding. On some occasions, detectable amounts of the fission product ^{131}I have been found in *F. vesiculosus* near Barsebäck. We have no definite proof that this radionuclide actually comes from Barsebäck. One explanation of the presence of ^{131}I could be that it is released from the hospitals in Malmö or Lund in connection with thyroid therapy (Mattsson *et al.*, 1977; Erlandsson & Mattsson, 1978). No other short-lived radionuclides normally present in fresh fallout from nuclear weapon tests have been detected at the same time. This indicates that the ^{131}I observed was not connected with fallout from recent bomb tests. The concentration of ^{137}Cs in *F. vesiculosus* near Barsebäck shows values which are normal for the Swedish west coast and the operation of the power station has not as yet resulted in measurable contamination of the marine environment by ^{137}Cs (details to be published later). The concentrations of $^{239+240}\text{Pu}$ and ^{241}Am in *F. vesiculosus* from the Barsebäck area show normal fallout levels.

Comparison of the ^{60}Co -activity concentrations north and south of Barsebäck

During the period June 1976 to August 1978, the ratio between the ^{60}Co -activity concentrations at Vikhög (2.7 km south of Barsebäck) and at our sampling location,

2.9 km north of the plant, was 0.18 ± 0.02 . Correcting for the slight difference in distance gives a 'south/total' ratio for the released activity of 0.13 ± 0.02 and a 'north/total' ratio of 0.87 ± 0.02 .

These values are in good agreement with the results of hydrological measurements (SMHI, 1978) made around Barsebäck which show that the current at 4 m depth in 1977 was directed northwards 83 % of the time and southwards for the remainder. In 1976, the corresponding figures were 77 % and 23 %, respectively.

Variation of the activation product concentration along the Swedish west coast north of Barsebäck

Figure 4 shows the activity concentrations of ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn and $^{110}\text{Ag}^{\text{m}}$ in *F. vesiculosus* at various distances north of Barsebäck in June 1977, shortly before the 1977 refuelling and service to reactor B1. Figure 5 shows the situation for ^{60}Co at three different times—June 1977, April 1978 (before the service of B1) and July 1978 (after the service). No significant change in the relative concentration gradient can be

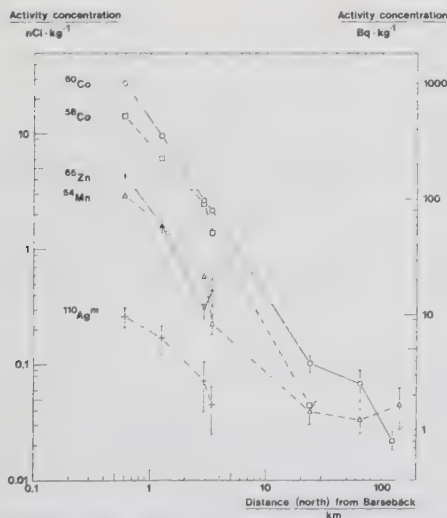


Fig. 4. Variation in activity concentration in *Fucus vesiculosus* by distance northwards from Barsebäck on 12 June 1977.

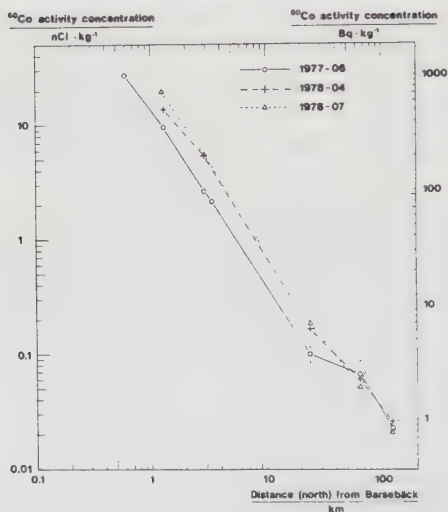


Fig. 5. Variation of ^{60}Co -activity concentration in *Fucus vesiculosus* by distance northwards from Barsebäck at three different times. 12 June 1977, 27 April 1978, and 5 July 1978.

observed, in spite of the activity concentration being more than doubled between June 1977 and April 1978.

The results for all five radionuclides indicate that the variation in activity concentration with distance at different times may be described by a power function of the form:

$$C(z) = \alpha \cdot z^{-\beta} \quad (2)$$

where $C(z)$ is the activity concentration at a distance, z , north of Barsebäck (km) and α and β are constants.

Table 1 lists the values of the constants α and β calculated for the different radionuclides on each of the three different sampling occasions. No significant change in the value of β can be observed among the radionuclides ^{60}Co , ^{58}Co , ^{65}Zn and ^{54}Mn at different times of collection; its mean value ($\pm$ SD) is 1.4 ± 0.1 . For ^{51}Cr and $^{110}\text{Ag}^m$ the information is very limited. However, the β -value for these radionuclides seems to be significantly lower than 1.4.

The disturbance of the activity gradients studied by release from the Ringhals

TABLE 1

VALUES OF THE CONSTANTS α AND β IN EQUATION (2), $C(z) = \alpha \cdot z^{-\beta}$, DESCRIBING THE RELATIONSHIP BETWEEN ACTIVITY CONCENTRATION $C(z)$ AND DISTANCE (z) FROM Barsebäck. THE STANDARD ERROR OF β , INDICATED IN THE TABLE, HAS BEEN CALCULATED AS:

$$\sqrt{\frac{\left(\sum z_i^2 - \frac{1}{N}(\sum z_i)^2\right)\left(\sum (C(z_i))^2 - \frac{1}{N}(\sum C(z_i))^2\right) - \left(\sum C(z_i) \cdot z_i - \frac{1}{N}\sum C(z_i)\sum z_i\right)^2}{(N-2)\left(\sum z_i^2 - \frac{1}{N}(\sum z_i)^2\right)}}$$

WHERE N IS THE NUMBER OF MEASUREMENT STATIONS BETWEEN 0.6 AND 143 km NORTH OF Barsebäck

Radionuclide	12 June 1977		27 April 1978		5 July 1978	
	α	β	α	β	α	β
⁶⁰ Co	11.9	1.33 ± 0.06	13.5	1.29 ± 0.08	26.8	1.50 ± 0.04
⁵⁸ Co	7.86	1.25 ± 0.16	1.12	1.41 ± 0.22	8.13	1.59 ± 0.02
⁶⁵ Zn	2.08	1.48 ± 0.22	—	—	4.14	1.72 ± 0.06
⁵⁴ Mn	1.74	1.33 ± 0.31	0.83	1.29 ± 0.16	1.70	1.37 ± 0.03
¹¹⁰ Ag ^m	0.17	0.78 ± 0.02	—	—	—	—
⁵¹ Cr	—	—	0.70	1.18 ± 0.05	—	—

nuclear power station (compare Fig. 1) is insignificant. Even if a considerable part of the ⁶⁰Co-activity concentration measured in *F. vesiculosus* from the sampling station 125 km north of Barsebäck (63 km south of Ringhals) did come from the latter, its contribution cannot have significantly influenced the concentration values for the stations closer to Barsebäck and hence cannot have altered the β -values in Table 1. The conformity of variations in activity concentration with distance north of Barsebäck at different times and for different radionuclides is remarkable and may indicate that the overall distribution of activity is governed by hydrodynamic conditions which are constant if averaged over months.

It is of some interest to discuss the numerical value of β obtained: the liquid radioactive waste is thoroughly mixed with the cooling water and brought out into the Öresund. The speed of the dominating north-bound current is, for most of the time, of the order of 0.2–0.3 m/s, which value is enough to transport released activity about 2000–5000 m downstream during the time required for a single injection (3–5 h). On its way downstream, the volume of contaminated water will expand due to turbulence and diffusion. If the expansion rate were constant in time and direction, the volume of contaminated water would be proportional to r^3 , where r is the 'mean' radius of the volume.

Expansion is, however, limited upwards by the water surface and downwards by the sea-bottom and sometimes by a deep leap layer of salt water. Thus, the vertical expansion is limited to approximately 10–15 m. This limit is probably reached very soon after the release, allowing the succeeding expansion to proceed in two dimensions only. In that case the volume of contaminated water would be proportional to r^2 and the activity concentration proportional to r^{-2} . If the speed of the current is temporally constant, the distance, z , travelled by the volume of

contaminated water will be proportional to time and hence to the 'mean' radius, r . In that case, the activity concentration in water or *F. vesiculosus* would be proportional to z^{-2} .

If, however, the expansion rate decreases with time, the exponent β should be less than 2, in accordance with our results.

SUMMARY AND CONCLUSION

The brown seaweed *Fucus vesiculosus* has been shown to be a very convenient bioindicator for mapping the local and distant spreads in the marine environment of the activation products ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn and $^{110}\text{Ag}^m$ from a boiling water reactor. The concentrations of these radionuclides were so low that it would have been very difficult to carry out a sufficiently large number of analyses in order to obtain the spatial distribution of the activity in water.

A comparison of the activity released from Barsebäck and the activity concentration in *F. vesiculosus* at a locality 2.9 km north of the discharge pipe shows that the different activation products studied can be ordered in the following sequence after the degree of uptake per activity unit released:

$$^{54}\text{Mn} \approx ^{58}\text{Co} \approx ^{60}\text{Co} \approx ^{65}\text{Zn} > ^{110}\text{Ag}^m > ^{51}\text{Cr}$$

The biological half-life of ^{58}Co (and ^{60}Co) in *F. vesiculosus* was found to be 60 ± 15 days. Approximately the same value was found for ^{54}Mn and ^{65}Zn .

The decrease in activity concentration, $C(z)$, with distance, z , north of Barsebäck can be described by a power function $C(z) = \alpha \cdot z^{-1.4}$, where z is given in kilometres and α is a constant. Our results show that, averaged over some time, 85–90% of the activity released is transported northwards and 10–15% southwards from Barsebäck.

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INTERNATIONAL SYMPOSIUM ON THE IMPACTS OF RADIONUCLIDE RELEASES INTO THE MARINE ENVIRONMENT

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RADIONUCLIDES IN FUCUS FROM INTER-SCANDINAVIAN WATERS.

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ABSTRACT

The brown algae *Fucus vesiculosus* and *Fucus serratus* were used to map the temporal and spatial variations in the distribution of activation products released from two nuclear power plants located on the Swedish west coast.

The *Fucus* plants well reflect variations in activity levels in the surrounding water. This property of *Fucus* together with knowledge of the amounts and time variations of the rate of released activity makes it possible to calculate uptake and retention parameters for these bioindicators. The uptake of the activation products per unit activity released was found to be of the same order for ^{60}Co , ^{58}Co , ^{54}Mn and ^{65}Zn but considerably less for $^{110}\text{Ag}^{\text{m}}$ and ^{51}Cr . Differences in the uptake in *Fucus* per unit activity released between the two areas were noted, and may be due both to differences in the dilution of the discharged activity and different physical and chemical form of the releases. The retention of these radionuclides in *Fucus* is believed not to be total; some biological elimination evidently occurs.

Concentration factors from water to *Fucus* were experimentally determined for ^{60}Co , ^{137}Cs and some natural and artificial actinides. These factors were found to be very high in the waters of the Swedish west coast. The ratio of $^{228}\text{Th}/^{228}\text{Ra}$ was used to estimate the age of the *Fucus* plants at the time of collection.

RADIONUCLIDES IN FUCUS FROM INTER-SCANDINAVIAN WATERS.

INTRODUCTION

The brown algae *Fucus vesiculosus* and *Fucus serratus* have been shown to be very suitable bioindicators for several radionuclides (1, 2, 3) as well as stable elements (4, 5, 6). To map the distribution of radionuclides in the marine environment and to reveal their transport it is very convenient to use such bioindicators which concentrate the substances since analytical procedures for seawater are very time-consuming. It is also of importance to study the activity concentration in algae because they are used in different kinds of food. The purpose of this work was to study the distribution of activation products released from the nuclear power plants at Barsebäck and Ringhals along the coasts of the inter-Scandinavian waters Öresund, Kattegatt and Skagerrack as well as the distribution of natural (Thorium and Uranium) and artificial (Plutonium and Americium) actinides.

MATERIAL AND METHODS

Samples of *Fucus* were collected at the Danish and Swedish coasts from water depths between 0.5 and 1 meter. After drying in air at roomtemperature for 2-3 days the samples were ground and packed in plastic containers. Gamma spectrometry was carried out using Ge(Li)-detectors with counting times between 5 and 100 hours. After measurements the samples were dried at 105°C for 24 hours to get the reference weight.

The Ge(Li)-detectors were carefully calibrated using samples of different geometries and densities containing accurately known activities of ^{152}Eu , ^{22}Na and ^{57}Co . Water samples with volumes between 100 and 240 liters were taken at some of the *Fucus* fields. ^{57}Co and ^{134}Cs were added as yield monitors. The pH was changed to pH=12 with NaOH to precipitate cobalt and actinides. The precipitate was recovered, dissolved and reprecipitated with ammonia. This second precipitate was centrifuged and dried before Ge(Li)-measurement. The cesium was precipitated with ammonium-molybdophosphate (AMP).

The actinides were separated radiochemically using ^{242}Pu , ^{243}Am , ^{232}U and ^{229}Th as radiochemical yield determinants. The alpha activity of the samples electrodeposited on to stainless steeldiscs were measured with surface barrier detectors for 1-20 days.

RESULTS AND DISCUSSION

A. Concentration of activation products in *Fucus*

From studies in the Barsebäck area it has been shown (1, 2, 3) that the time variation of the concentration of activation products in Fucus reflects well the increase in the activity discharged during the overhaul periods. The effective half life of cobalt, zinc and magnese in Fucus was found to be around two months in summertime (1) .

Figure 1 shows the spreading pattern of ^{60}Co as measured in Fucus from a number of sampling stations around the Swedish west coast and the Danish east coast as well as from islands in Kattegatt and Öresund. The map shows the highest activity concentration in the Barsebäck area with concentrations up to 3000 Bq per kg dry weight of Fucus. The concentration decreases very rapidly with distance from the plant and the minimum value along the coast is around a factor of 1000 lower. In the Ringhals area lower activity concentration levels are reached although the released activity from Ringhals is of the same order of magnitude as that from Barsebäck. These differences will be discussed in detail in later sections.

The map also shows that ^{60}Co from Barsebäck follows the north bound water stream up to Öresund and Kattegatt. Comparatively high activity concentration values are also found just north of Copenhagen which could indicate a whirl-pool south of the island of Ven before the stream continues mainly along the Swedish west coast.

Earlier it was shown (1, 2, 3) that the spread of radionuclides from the power plant in Barsebäck could be described by a power function of form $C(z) = \alpha z^{-\beta}$, where $C(z)$ is the activity concentration at distance z and α and β are constants. The value of β was found to be 1.4 for the radionuclides ^{60}Co , ^{58}Co , ^{65}Zn and ^{54}Mn somewhat lower, 0.8 for $^{110}\text{Ag}^m$ and still lower for ^{131}I in the Barsebäck area. The results for the Ringhals area give somewhat lower β -values. The vertical turbulation at Ringhals is greater than that at Barsebäck depending on differences in the water depths of the outlets (surface Barsebäck, bottom Ringhals). The β -value might then to some extent also be dependent on this difference in vertical dilution. At Ringhals the lower uptake in Fucus at a given distance indicates a better dilution of the discharged activity which is in accordance with oceanographic reports (7) .

B. Mathematical description of uptake and retention in Fucus

Since September 1976 the long-term variation of the activity concentration of activation products in Fucus has been studied at many locations along the Swedish west coast. Before this date the activity released monthly was around a factor 100 lower than after the first overhaul period at Barsebäck in the summer of 1976.

Using information on the controlled releases of radionuclides

from the power plants in Barsebäck and Ringhals (8) it has been possible to study differences in the uptake and retention in Fucus of various activation products. These studies have shown a similar uptake and retention pattern for ^{60}Co , ^{58}Co , ^{65}Zn and ^{54}Mn but considerably less uptake for $^{110}\text{Ag}^{\text{m}}$ and still less for ^{51}Cr per unit activity released (1, 2, 3).

Most of the calculations were carried out using ^{60}Co as the tracer radionuclide. The reason for this is that the released ^{60}Co -activity is much greater than that for the other activation products, thus resulting in less statistical error in the activity concentration values in Fucus. Two different methods are described in the following paragraphs.

Assuming that the concentration of ^{60}Co in the water surrounding the seaweed varies proportionally with the discharge rate $A(t)$, the time-dependent ^{60}Co -activity concentration $C(t)$ in Fucus may be expressed as:

$$\frac{dC(t)}{dt} = k \cdot A(t) - (1 + \lambda) C(t) \quad (1)$$

where k is a constant, describing the increase in activity concentration in Fucus after a release of unit activity (kg^{-1}), and where the constant 1 (d^{-1}) describes the apparent elimination and is probably due to both growth dilution and biological elimination. The last parameter λ is the physical decay rate constant (d^{-1}).

Using a known starting value of the activity concentration $C(0)$, the equation is numerically integrated using different combinations of k and 1 -values until the best fit to the experimental points is found.

Another method to quantify the uptake is to use a decay-corrected transfer factor DTF_m . This factor is calculated as the ratio between the activity concentration in Fucus at a given location ($\text{Bq} \cdot \text{kg}^{-1}$) and the discharge (Bq) decay-corrected to the sampling time during m preceding months, or in other words, the fraction of m months discharge found in one kg of the sample. This factor is comparable to the above mentioned k -value. The number of months in the calculation, m , is chosen as the number of months for which DTF_m -values for the short-lived ^{58}Co equals that of the relatively long-lived ^{60}Co .

This period of time, m , is also given as the integration time, as the radiocobalt concentration actually measured in Fucus probably reflects the discharges during the preceding m months. Further details on the DTF_m -calculations have been published elsewhere (9, 10).

In figure 2, the time-variation of the (measured) activity concentration in Fucus at a location 2.9 km north of the Barsebäck

discharge pipe is shown. The monthly discharge rate of ^{60}Co is also shown, as well as the calculated activity concentration using equation (1). The integration is carried out in three periods, starting and ending in summer. The best values of k and l for each period as well as the mean DTF_m -value for this period are given in table I.

The constants k and DTF_m are, of course, dependent on the distance from the outlet following the power function $z^{-1.4}$ mentioned above.

It is interesting to note the similarity between the k and DTF_m -values. This similarity repeats itself when the different methods of calculation are applied to a very frequent sampling in May-June 1978 at the location 2.9 km north of Barsebäck. These calculations were based on daily releases and eight samplings over a period of two months, and have been published recently by Mattsson et al. (1). The values for k and l in this case were $2.1 \cdot 10^{-8} \text{ kg}^{-1}$ and $1.2 \cdot 10^{-2} \text{ d}^{-1}$, respectively. The DTF_m -values during this period were between $0.8 \cdot 10^{-8} \text{ kg}^{-1}$ and $2.0 \cdot 10^{-8} \text{ kg}^{-1}$.

The number of experimentally determined activity concentrations in Fucus during these three years are unfortunately too few to allow an estimation of seasonal variations of these three constants. It is evident that the DTF_m -values oscillate through the year showing the highest values in autumn and winter and the lowest values in spring and summer. However, during the three years that were covered, this pattern did not reproduce itself completely and furthermore the variations were nearly parallel to the discharge rate pattern, showing the lowest DTF_m -values after some months with low discharge rate (spring) and the highest values after periods with high discharge rates (autumn). This could, however, be explained by growth dilution during spring and summer. On the other hand, for stable zinc in Fucus spiralis van Weers (6) found a similar seasonal variation that could indicate that part of the oscillation might be valid. An extended study of the k -value using frequent collections over some years would show the true variations in uptake, since this quantity does not suffer from the effects of any form of elimination or dilution process.

C. Differences in the uptake of radionuclides between the two areas

Figure 3 shows the variation of ^{60}Co activity concentration in Fucus to the south and north of the nuclear power plants on two occasions in 1977. These results can be compared with the releases from the plants during the same year (figure 4).

The figures show that during June-July the concentration of ^{60}Co in Fucus from both areas was almost equal, but the releases before

and during the period are higher at Ringhals than at Barsebäck. In October-November the activity concentration of ^{60}Co in Fucus from Barsebäck was about five times higher than that of Ringhals despite the fact that the release at Ringhals was higher.

The conclusion must be that the discharged ^{60}Co activity during these periods was more efficiently transferred to Fucus at Barsebäck than at Ringhals.

The salinity of the surface water at Ringhals is about 20‰ and that at Barsebäck 10‰ (7) . However, an analysis of stable cobalt, shown in table II, imply that the differences in salinity are too small to cause a difference in the uptake.

Water currents in Öresund are generally northerly, affected by the outgoing current from the Baltic (7) . Consequently, the release from the nuclear power plant at Barsebäck mainly flow northwards as well and is pressed by the currents against the shallow shore. The dominant direction of the current at Ringhals is also northerly (11) , but the water at the coast is relatively deep and the coast is exposed to the open sea. The releases are probably thus more diluted at Ringhals than at Barsebäck resulting in less transferred activity to the Fucus plants. However, for ^{65}Zn the picture is different.

The constant k in the mathematical model for the uptake of radionuclides in Fucus presented previously has been calculated for ^{65}Zn at the two sites and corrected so that it is valid for the same distance from the nuclear power plants. To ensure reliable data, calculations were made when distinct releases and samplings with dense frequencies were obtained. Table III shows the k -values at a distance of 2.9 km from the plants at two occasions.

The uptake constant k for ^{65}Zn is evidently of the same order of magnitude in both areas.

These differences between the transfer of discharged ^{65}Zn and ^{60}Co show that the differences between the sites are not only caused by differences in dilution. Probably the physical and chemical conditions of the released radionuclides are different between the two power plants. For instance the ^{65}Zn releases from Ringhals during the period mentioned in table III were dominated by particulate activity, which might have caused a surface contamination. Thus, the differences observed between the two power plants might be caused by differences in the physical and chemical state of the nuclides released.

D. Activation products in water and Fucus

In May 1980 two water samples of 240 kg were taken at two

different locations in the Ringhals area. Gamma-spectrometry of the precipitate showed the ^{60}Co , ^{58}Co and ^{137}Cs activities given in table IV. This table also gives the activity concentration values in Fucus at the same time and location as well as the concentration factors.

The concentration factors are surprisingly high, yet still in accordance with earlier results from the Barsebäck area $((2 \pm 1) \cdot 10^3)$ (3). It must strongly be stressed that, owing to the pulsed release pattern, these data could be an overestimation of the true concentration factor if the water sample is not representative of the period during which the Fucus plant was exposed to the radio-nuclides. Thus, except for ^{137}Cs these concentration factors should be used with great caution.

E. Actinides in water and Fucus

Fucus is also an excellent bioindicator for actinide elements, especially the transuranic elements.

The samples collected for studies of activation products were used also for this purpose. In table V the average results for 1978 and 1979 are given. The values for water still refer to unfiltered water.

The authors have not observed any influence of transuranics from European reprocessing facilities. Thus, the Plutonium and Americium detected comes entirely from global fall-out.

The results from water are in agreement with those of Murray et al. (12) in adjacent areas and the values for Fucus are close to those reported by Miettinen (13) and Bojanowski (14) from the Baltic.

The concentration factors for Plutonium and Americium are very high. Recently Hodge et al. (15) showed that significant uptake in particulate form takes place by coastal marine organisms. Plutonium and Americium are expected to be present predominantly in particulate form in the specific area studied.

Of the Thorium isotopes ^{228}Th differs remarkably from the others. ^{228}Th ($T_{1/2} = 1.91 \text{ a}$) is a daughter product of ^{228}Ra . The $^{228}\text{Th}/^{228}\text{Ra}$ activity ratio in the samples is about 0.3. Assuming that the ^{228}Ra concentration is constant expressed as $\text{Bq} \cdot \text{kg}^{-1}$ and by calculating the in vivo build up of ^{228}Th and correcting for uptake of ^{228}Th from the water by use of table V the mass average age of the Fucus samples could be estimated to be 11-12 months.

This technique could generally be used to estimate the age of marine organisms, thus improving their value as bioindicators.

SUMMARY AND CONCLUSION

The brown seaweed *Fucus vesiculosus* and *Fucus serratus* have proved to be excellent bioindicators for mapping the spreading of activity released into the marine environment. The *Fucus* plants respond very rapidly to fluctuations in activity levels in the surrounding water. Parameters describing uptake per unit activity released have been calculated in two different ways, giving almost equal results. The uptake parameters are, however, dependent on both the site of collection and the physical and chemical form of the releases. The concentration factors for activation products are found to be very high in the waters of the Swedish west coast, and do not differ much between the two areas studied. The sampling technique for water can, however, be improved giving more representative samples with regard to the time during which the *Fucus* plants have been exposed. The concentration factors for actinides are also found to be very high in these areas.

These techniques for analysis could also be applied to the amounts of fission products that lately have started to increase in the vicinity of Barsebäck. This would substantially contribute to the exploration of many white areas on the radioecological map of the inter-Scandinavian waters.

ACKNOWLEDGEMENT

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TABLE I

Values for k and l in equation 1 giving the best fit to the experimental points. Mean values of DTF_m during these periods of time. Biological half-time is calculated from the value of the constant l .

Period	$k(kg^{-1})$	$DTF_m(kg^{-1})$	$l(d^{-1})$	$T_{1/2}(d)$
Sept.1976-July 1977	$4.8 \cdot 10^{-8}$	-	$0.7 \cdot 10^{-2}$	100
July 1977-July 1978	$2.4 \cdot 10^{-8}$	$2.5 \cdot 10^{-8}$	$1.2 \cdot 10^{-2}$	60
July 1978-Aug. 1979	$7.2 \cdot 10^{-8}$	$4.0 \cdot 10^{-8}$	$0.8 \cdot 10^{-2}$	85

TABLE II

Contents of stable cobalt in Fucus (dry weight) and water at Ringhals and Barsebäck in spring 1980.

	Fucus($mg \cdot kg^{-1}$)	Water ($\mu g \cdot l^{-1}$)
Ringhals	1.9 ± 0.4	0.05 ± 0.03
Barsebäck	1.9 ± 0.2	0.05 ± 0.03

TABLE III

k-values (kg^{-1}) (equation 1) for ^{65}Zn at Ringhals and Barsebäck 2.9 km from the discharge pipes.

	$k(\text{kg}^{-1})$
Ringhals, May-August, 1979	$4.2 \cdot 10^{-8}$
Barsebäck, May-July, 1978	$2.0 \cdot 10^{-8}$

TABLE IV

Activity concentration in Fucus and water at Ringhals for ^{60}Co , ^{58}Co and ^{137}Cs and the corresponding concentration factors (CF). All figures for Fucus are on dry weight basis.

Sample	$^{60}\text{Co}(\text{Bq} \cdot \text{kg}^{-1})$	$^{58}\text{Co}(\text{Bq} \cdot \text{kg}^{-1})$	$^{137}\text{Cs}(\text{Bq} \cdot \text{kg}^{-1})$
Fucus 1	78 ± 3	73 ± 5	7.4 ± 0.4
Water 1	$(5.6 \pm 0.7) \cdot 10^{-3}$	$(1.1 \pm 0.4) \cdot 10^{-3}$	$(78 \pm 4) \cdot 10^{-3}$
CF*	$14\ 000 \pm 3\ 000$	$66\ 000 \pm 28\ 000$	95 ± 10
Fucus 2	111 ± 4	45 ± 5	7.4 ± 0.4
Water 2	$(1.5 \pm 0.15) \cdot 10^{-3}$	$(0.8 \pm 0.2) \cdot 10^{-3}$	$(93 \pm 5) \cdot 10^{-3}$
CF*	$75\ 000 \pm 10\ 000$	$56\ 000 \pm 25\ 000$	80 ± 10

* See text for explanation

TABLE V

Actinides in water and Fucus

	^{228}Th	^{230}Th	^{232}Th	^{234}U	^{235}U	^{238}U	$^{239+240}\text{Pu}$	^{241}Am
Water ($\mu\text{Bq kg}^{-1}$)								
Surface	450	270	70	18100	650	15900	14	3
Bottom 19m							40	7
Fucus (mBq kg^{-1}) (dry)	5000	400	90	12700	450	11100	190	53
CF (dry)	11000	1300	1300	700	700	700	13500	18000

FIGURE CAPTIONS

Figure 1: The spreading pattern of ^{60}Co from the two nuclear power plants in Ringhals and Barsebäck given as activity concentration (Bq kg^{-1}) in *Fucus vesiculosus* on dry weight basis.

● = April 1979. ○ = December 1978 corrected to be valid for April 1979.

Figure 2: Time-variation of measured and calculated ^{60}Co activity concentrations in *Fucus vesiculosus* at a location 2.9 km north of the discharge pipe in Barsebäck and the ^{60}Co activity discharge rate from this power plant.

Figure 3: Spatial variation of ^{60}Co activity concentration in *Fucus vesiculosus* to the south and north of the two power plants Ringhals and Barsebäck on two occasions in 1977.

Figure 4: Monthly discharge rate of ^{60}Co from the two power plants Barsebäck and Ringhals during 1977.

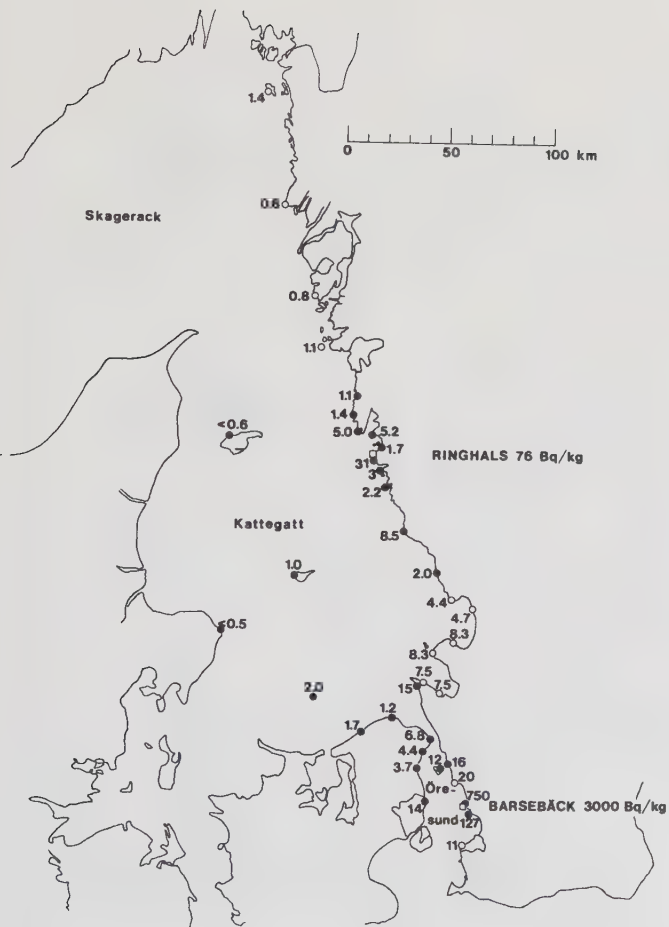


FIGURE 1

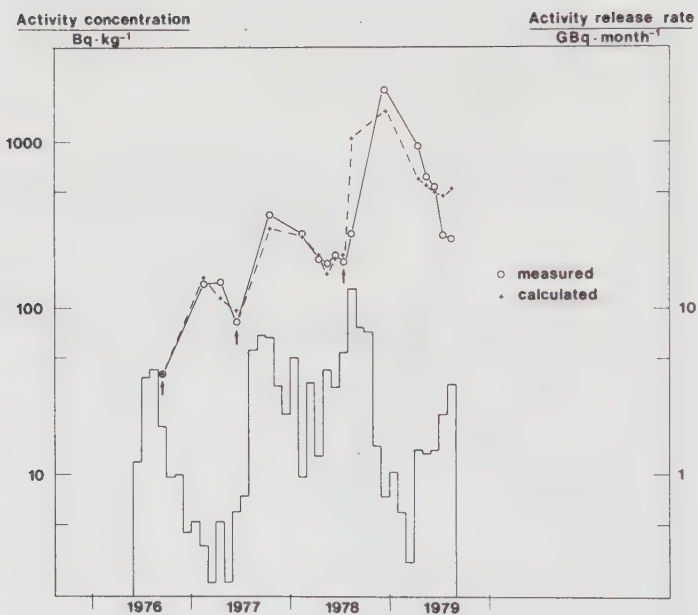


FIGURE 2

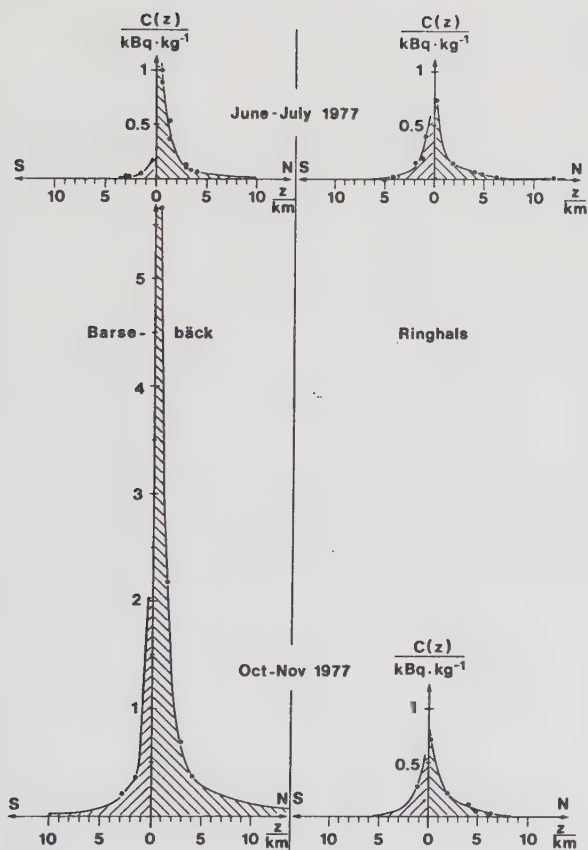


FIGURE 3

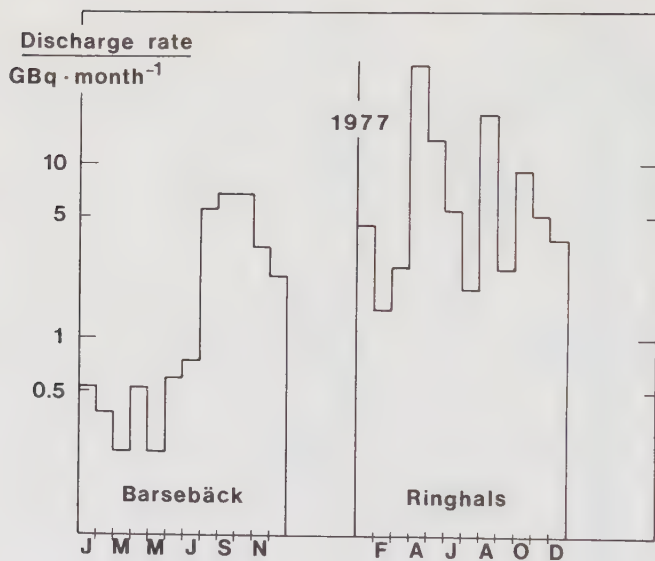


FIGURE 4

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CODEN:LUMEDW/(MERI-3025)/1-19/(1981)

Radioecological studies of activation products released from
a nuclear power plant into the marine environment.

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INTRODUCTION

The handling and processing of nuclear fuel and waste give rise throughout the fuel cycle to releases of radioactive substances into the environment. An important source at present and probably the most important in the future for the irradiation of man is the marine environment.

We have therefore considered it of great importance to study the transport of radionuclides released under controlled conditions from a nuclear power plant into the marine environment. This applies to both the radionuclide content in water and its concentration along food-chains of interest with respect to estimating the dose commitment to the exposed population.

The discharged radioactivity is, however, dispersed into enormous volumes of water. It is therefore desirable to find a bioindicator which concentrates the substances but still, to a certain degree, describes the radionuclide concentration in its immediate surroundings. We have found species of the brown seaweed Fucus to be very suitable bioindicators for such studies (1).

The source of the discharged radioactivity is the Barsebäck nuclear power plant which is located on the Öresund sound between Denmark and Sweden. It consists of two boiling water reactors, each of 1700 MW thermal effect. The first reactor (B1) became operational in 1975, and the second (B2) in 1977 (cf. figure 1). The release of radionuclides, mainly activation products, is during normal operation, due to high filtering and waste containment capacity, quite low. During the summer period, when annual overhaul and partial refuelling takes place, the discharge is much higher. This gives a pulsed pattern of released radionuclides, which is fortunate for our studies, since it simplifies the interpretation and mathematical treatment of data regarding the time variations of concentration, uptake and retention of the radionuclides.

Our studies of the radionuclide content in Fucus started in 1967; being at that time mainly studies of the long term variation of ^{137}Cs activity concentration. These studies were carried out at a considerable distance (140 km north) from the power plant. In the summer of 1976 we for the first time found traces of ^{60}Co in

Fucus from this collecting station. From that time on, we expanded our programme to regular collection at eight sampling stations.

MATERIAL AND METHODS

Samples of Fucus vesiculosus, F. serratus, Ascophyllum nodosum and Cladophora glomerata, firmly rooted on heavy stones were collected from water depths between 0.5 and 1 m along the coast. Samples of the crustaceans Idothea, I. baltica, I. viridis and I. granulosa, as well as Gammarus found on the Fucus plants were sorted out for analysis. A number of samples of the mussel Mytilus edulis as well as a limited number of sediment profiles were also taken. After drying in air at room temperature for 2-3 days the samples were ground and packed in plastic containers of 5 or 180 ml volume. The measurements were carried out using Ge(Li)-detectors of volume 46-100 cm³ with counting time between 5 and 100 hours. The detector efficiencies were determined carefully using samples of different densities containing accurately known activities of ¹⁵²Eu, ⁵⁷Co and ²²Na.

Water samples with volumes between 80 and 170 liters were taken at the Fucus fields 2.9 km NNE of the power plant. ⁵⁷Co was added as a yield determinator and NaOH was then added to pH=12. The precipitate ($\approx$ 5 l) was the next day dissolved in HCl and co-precipitated with 200 mg Fe as carrier. The pH was adjusted to 12 with ammonia. This precipitation was centrifuged and dried before measurement.

RESULTS AND DISCUSSION

A. Activation products in Fucus

The location where collection has most frequently been made is situated 2.9 km north of the discharge pipe. Figure 1 gives the activity concentrations (dry weight) for ⁶⁰Co ($T_{1/2}$ = 5.26 a), ⁵⁸Co (71.3 d), ⁵⁴Mn (303 d) and ⁶⁵Zn (245 d) in Fucus v. collected at this location between 1976 and 1980. On the abscissae, different operational data for the plant are given. We have also, on occasion, detected ^{110m}Ag (255 d), ⁵⁷Co (270 d), ⁵¹Cr (27.8 d) and ¹³¹I (8.05 d).

The activation products shown in figure 1 all follow a similar pattern, and clearly reflect the increase in discharged activity during the overhaul periods.

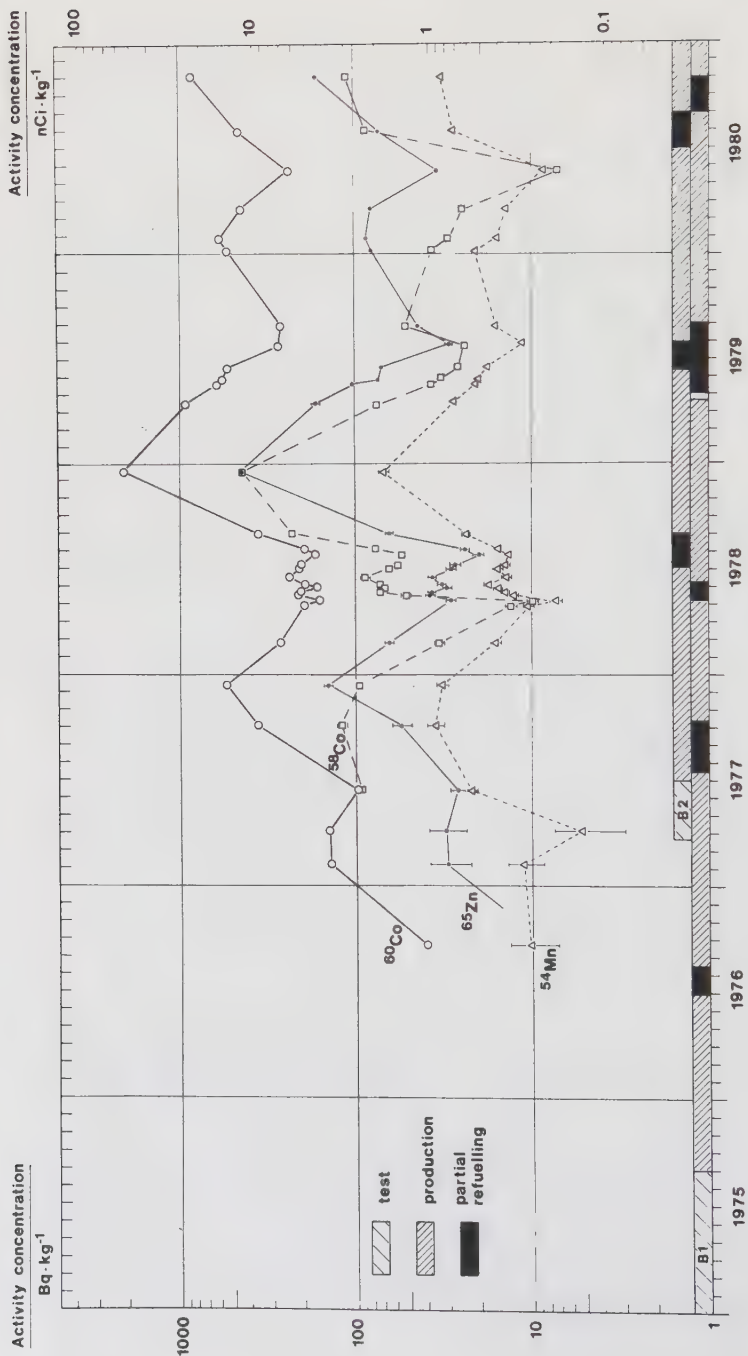


Figure 1. Time variation of the concentration of activation products in *Fucus v. 2.9 km* north of the Barsebäck discharge pipe

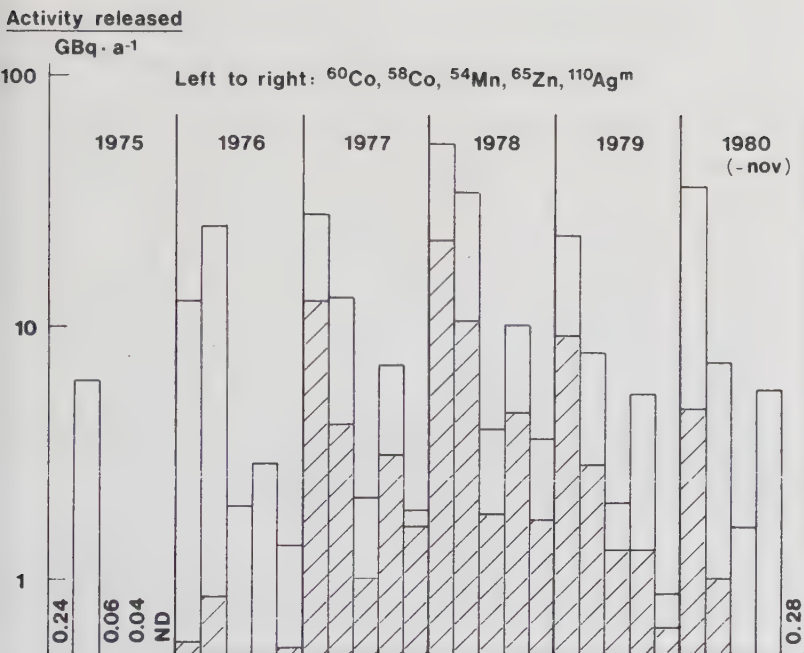


Figure 2. Annual discharge rates of activation products from Barsebäck nuclear power plant into the Öresund. The shaded parts correspond to activity that can be absorbed on a 45 µm Millipore filter.

From these results, we could establish that the increase in activity concentration in Fucus v. per unit activity released from the power plant was about the same for these radionuclides (1). Furthermore, from the very frequent collection in May-June 1978 we could determine the biological half-life for these nuclides to be (60 ± 15) days (1). For this purpose we were supplied with data on released activity by the staff of the power plant (2). The half-lives for the cobalt isotopes are in good agreement with laboratory experiments (3). For ^{65}Zn , however, our results indicate a lower retention than found in laboratory experiments. (4,5)

In figure 2 the annual discharge rates (from 1975 until November 1980) of the activation products are shown. In 1975 and 1976, when the first reactor was tested and started and ^{60}Co , due to its long half-life, not had reached high activity values, ^{58}Co dominated the discharges. After 1976, the radionuclides can be ranked, in terms of activity discharge, as: $^{60}\text{Co} > ^{58}\text{Co} > ^{65}\text{Zn} > ^{54}\text{Mn} > ^{110}\text{Ag}^{\text{m}}$. This order has also been found in the Fucus plants (cf figure 1).

The yearly activity discharge rate has, for all nuclides studied, a marked maximum in 1978, which also is reflected by the Fucus plants. In 1980, however, the ^{60}Co activity discharges again increased. The Fucus plants again responded to the increased discharge rate, as can be seen in figure 1. The activity content of the Fucus plants of the other activation products also increased in autumn 1980, due to the fact that most of the activity is discharged during the second half of the year. The reason for the increased activities was probably (6) that a valve in a system for flushing of the magnetite filters in reactor 1 was leaking. The general tendency is, except for this incident, that the activity discharged is being reduced after 1978. This has been accomplished by using a waste handling system giving more efficient filtering and evaporation of the liquid waste, thus reducing the volumes that are to be stored in the waste tanks (6).

It must be pointed out that the activity being discharged to the environment is extremely low compared to the limits allowed by Swedish regulations. In 1980, the activity released was, for beta + gamma emitters about 0.5% of allowed limits. (The absolute activity values were 60 GBq compared to the allowed limit of 11 100 GBq.)

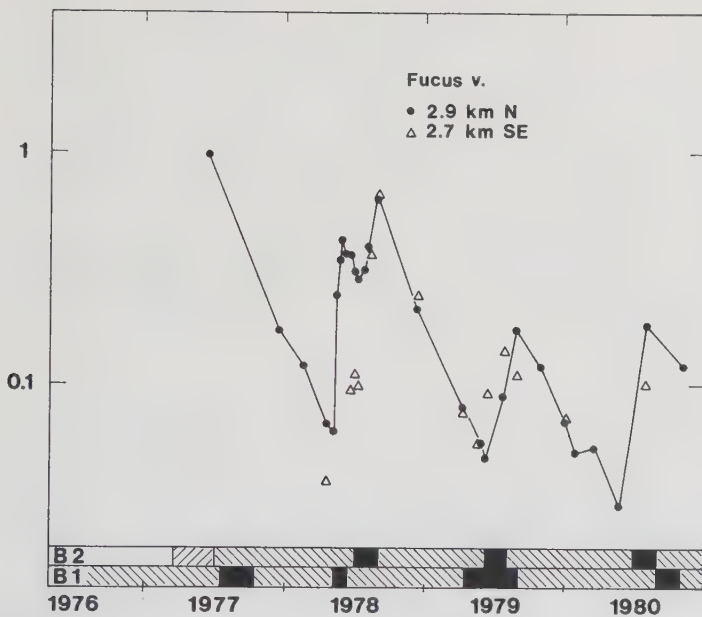
$\frac{^{58}\text{Co}}{^{60}\text{Co}}$ activity ratio


Figure 3. Time variation of $^{58}\text{Co}/^{60}\text{Co}$ ratio in *Fucus v.*

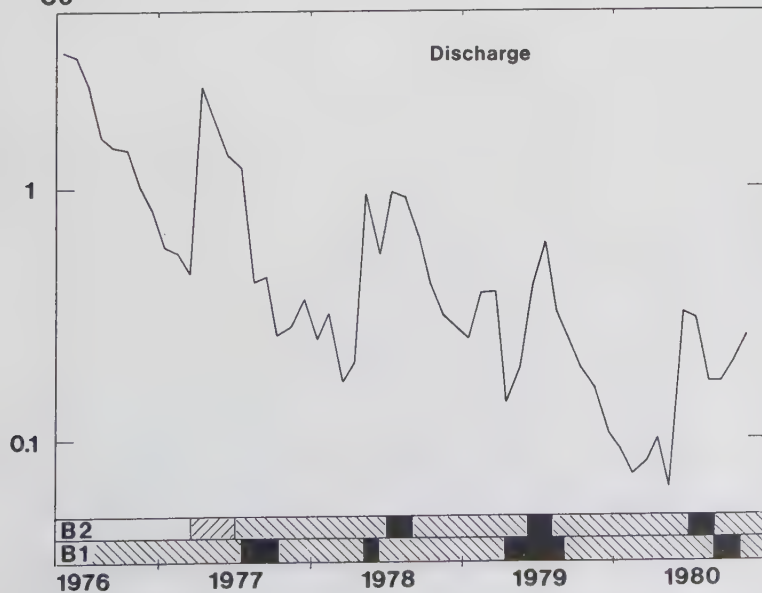
 $\frac{^{58}\text{Co}}{^{60}\text{Co}}$ activity ratio


Figure 4. Time variation of $^{58}\text{Co}/^{60}\text{Co}$ ratio in the liquid discharge.

We have also followed the ratio of $^{58}\text{Co}/^{60}\text{Co}$ activity concentrations in Fucus v. at the same sampling station 2.9 km north of the discharge pipe. This is presented in figure 3. From this figure it is obvious that the power plant releases ^{58}Co and other activation products mainly during the overhaul periods. This figure also clearly shows that the $^{58}\text{Co}/^{60}\text{Co}$ activity ratio is an excellent indicator of an increased discharge rate from the plant. The resolution in time is remarkably good although the interval between the two shutdowns in 1978 was only three weeks. The activity ratio still shows a decrease between the shutdowns.

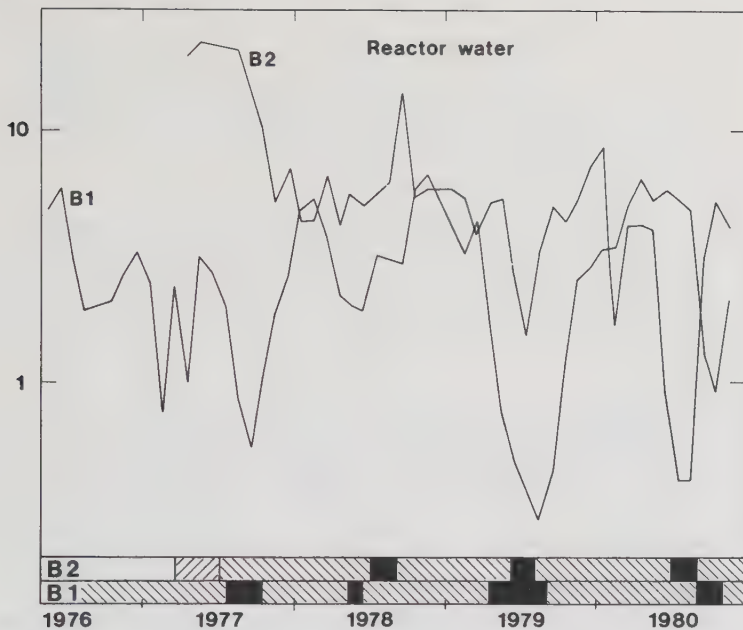
For comparison, the $^{58}\text{Co}/^{60}\text{Co}$ ratio in samples collected 2.7 km SE of the discharge pipe is also shown in figure 3 (triangles). The agreement is good, illustrating the stability of the distribution in time of the direction of the water current. This is in agreement with hydrological data (11).

In figure 4, the same ratio ($^{58}\text{Co}/^{60}\text{Co}$) in the liquid discharges are given. The curve at first surprisingly well seems to follow that for Fucus v. 2.9 km N of the discharge pipe. Linear regression of this ratio in discharge (x) and Fucus v. (y), however, gives the equation

$$y = 0.77 x - 0.005 \quad (r = 0.94) \quad (1)$$

The calculated factor 0.77 is mainly conducted by the ratio (of $^{58}\text{Co}/^{60}\text{Co}$) Fucus/discharge for the highest $^{58}\text{Co}/^{60}\text{Co}$ ratios. However, ^{58}Co is mainly produced in the reaction $^{58}\text{Ni}(n,p)^{58}\text{Co}$ and ^{60}Co in the reaction $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$. These cobalt isotopes could then be in different chemical states, thus initially giving them different chemical properties. The highest $^{58}\text{Co}/^{60}\text{Co}$ ratios are found in connection with the overhaul periods, and a major part of the ^{58}Co activity then released is recently produced. During the rest of the year, the ratio $^{58}\text{Co}/^{60}\text{Co}$ both in the discharge and in Fucus v. is lower and the ^{58}Co activity released has during some months (6) been stored in the waste tanks, where a change in chemical state and other chemical properties perhaps has taken place.

This assumption regarding ^{58}Co is supported by figure 5, where the $^{58}\text{Co}/^{60}\text{Co}$ ratio in the reactor water is shown. This ratio is during normal operation about 6, which is about 10-20 times higher

$\frac{^{58}\text{Co}}{^{60}\text{Co}}$ activity ratio


Activity concentration
Bq · kg⁻¹ (d. wt.)

Activity concentration
nCi · kg⁻¹ (d. wt.)

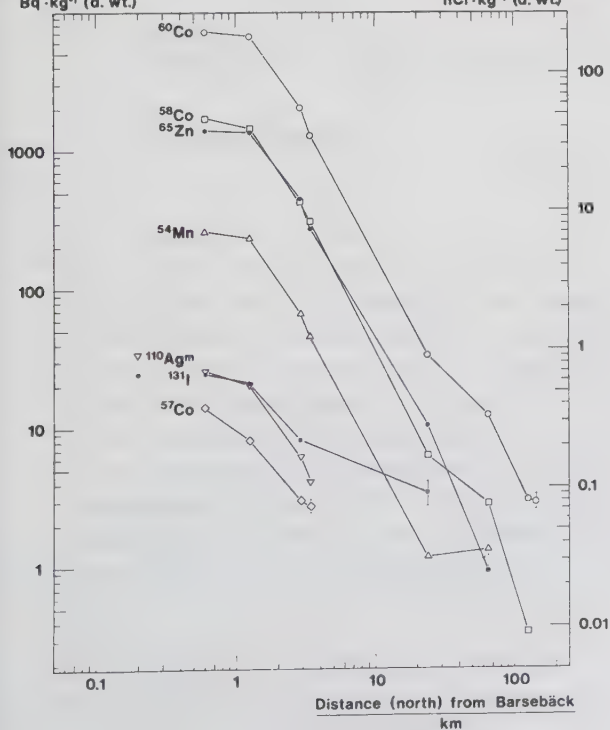


Figure 5 (above)

Time variation of
 $^{58}\text{Co}/^{60}\text{Co}$ ratio in
reactor water.

Figure 6. Variation
in activity concentration in *Fucus v.*
by distance north-
wards in December
1978.

than in the releases and in Fucus v.. Here the great difference could only be explained by different chemical behaviour of ^{58}Co and ^{60}Co in the ion-exchangers in the reactor water purification systems, thus indicating that their different origin is the explanation of their different behaviour.

The results from figure 3 and 4 also confirm the effect of the improved waste handling system. The $^{58}\text{Co}/^{60}\text{Co}$ ratio consistently decreases with time, which is in accordance with the reduced waste volumes and longer mean residence time in the waste tanks mentioned previously.

The more distant spread of activity has been investigated at eight collecting stations for Fucus v. along the Swedish west-coast. In figure 6, the variation with distance of the activity concentrations in Fucus v. for ^{60}Co , ^{58}Co , ^{54}Mn , ^{65}Zn , $^{110}\text{Ag}^{\text{m}}$ and ^{57}Co , all activation products, are given. This diagram is based on a collection made on the same day in December 1978.

As discussed in a previous article (1) it is clear that this distance dependence of the activity concentration gives a good fit to a power function of the form:

$$C(z) = \alpha \cdot z^{-\beta} \quad (2)$$

where $C(z)$ = activity concentration at distance z and α and β are constants.

A presentation of the time variation of the constants α and β is found in figures 7 and 8. The variation of the constant α for the different radionuclides is similar to the variation of the activity concentrations at 2.9 km north of the discharge pipe (cf. figure 1), thus indicating that the activity concentration in Fucus is, at all sampling stations, closely connected to the discharge rate from the plant. This indication is also supported by the similarity found in figures 2 and 7. In figure 8, the time variation of the constant β is given for the activation products. The value of this constant, as shown in figure 8, is nearly the same for all radionuclides, except $^{110}\text{Ag}^{\text{m}}$, which has a value of

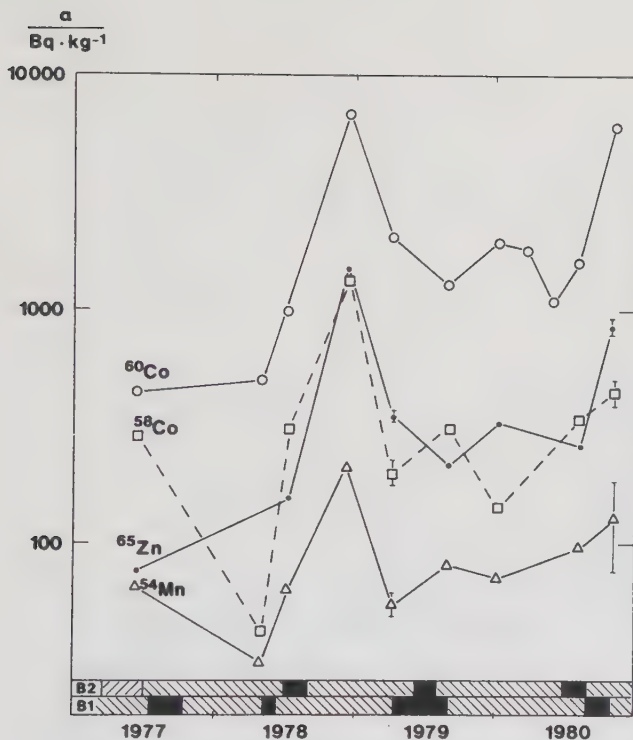


Figure 7. Time variation of the constant α (equation 2).

about 50% lower. A reason for this is probably that the silver released to the Öresund, due to its chemical properties, is behaving differently in the water compared to the other radionuclides. Differences in the uptake in *Fucus* has previously been reported (1).

In figure 8, a tendency can be seen that the β -values for the other four radionuclides show a similar time variation. Furthermore, the highest β -values seem to occur in connection with periods with high discharge rates. Since the current situation in the Öresund is very stable, this pattern of β -values could be linked to the dis-

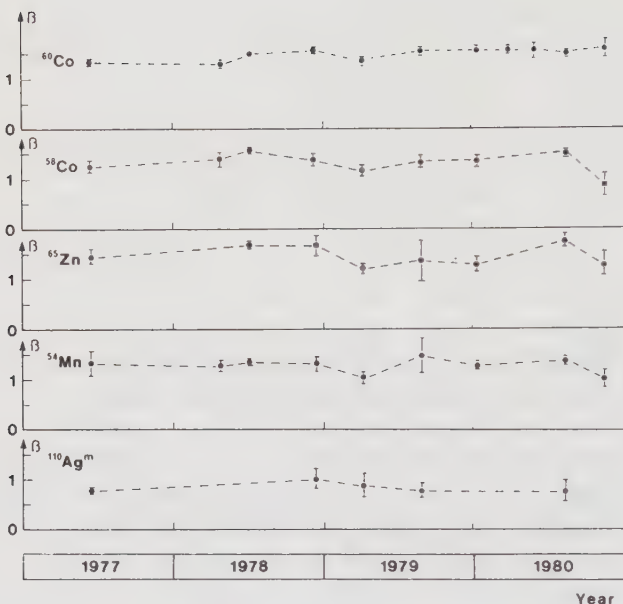


Figure 8. Time variation of the constant β (equation 2).

charge rate. The small variations of the constant β could accordingly be explained by there being a considerable time delay between the discharge and established equilibrium conditions between water, sediments and Fucus.

B. Studies of activation products in Idothea

Detectable concentrations of ^{60}Co were found in several samples of Idothea. Because of the limited sample weights (0.2-1 g dry weight) the analysis had normally to be restricted to ^{60}Co . Figure 9 shows a comparison of the ^{60}Co activity concentration in I. baltica and

Activity concentration

Activity concentration

Bq·kg⁻¹ (d. wt.)

nCi·kg⁻¹ (d. wt.)

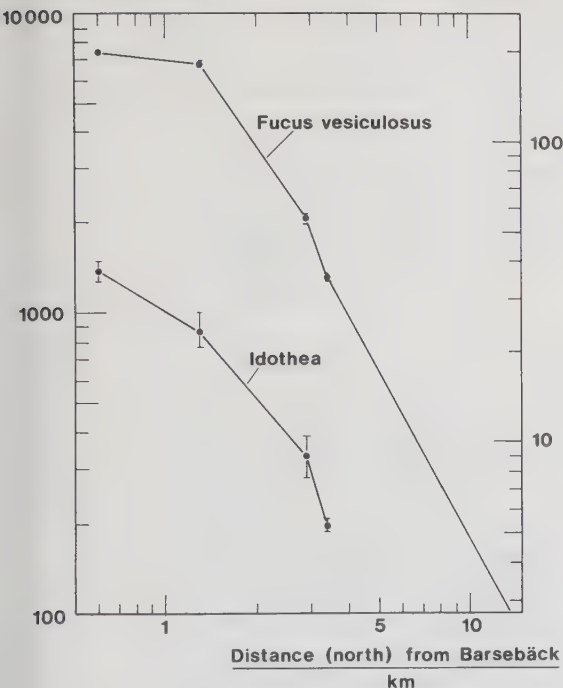


Figure 9 (left).

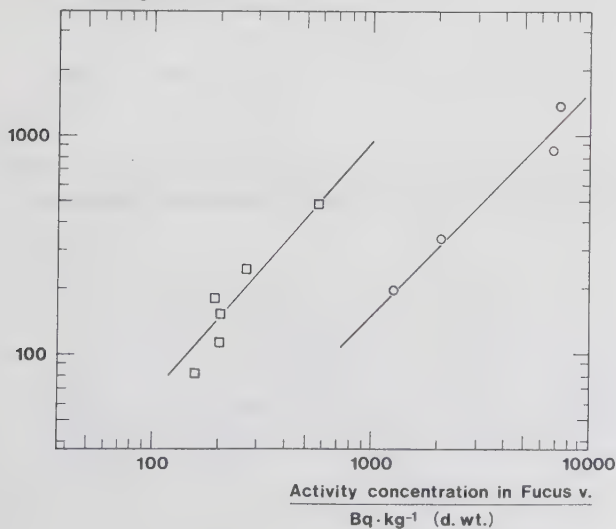
Variation in ⁶⁰Co activity concentration in *Fucus v.* and *Idothea Baltica* by distance northwards in Dec. 1978.

Figure 10 (below).

Relation between ⁶⁰Co activity concentrations in *I. baltica* and *Fucus v.* in summer (squares) and winter (circles).

Activity concentration in *Idothea*

Bq·kg⁻¹ (d. wt.)



Activity relative to Idothea B.

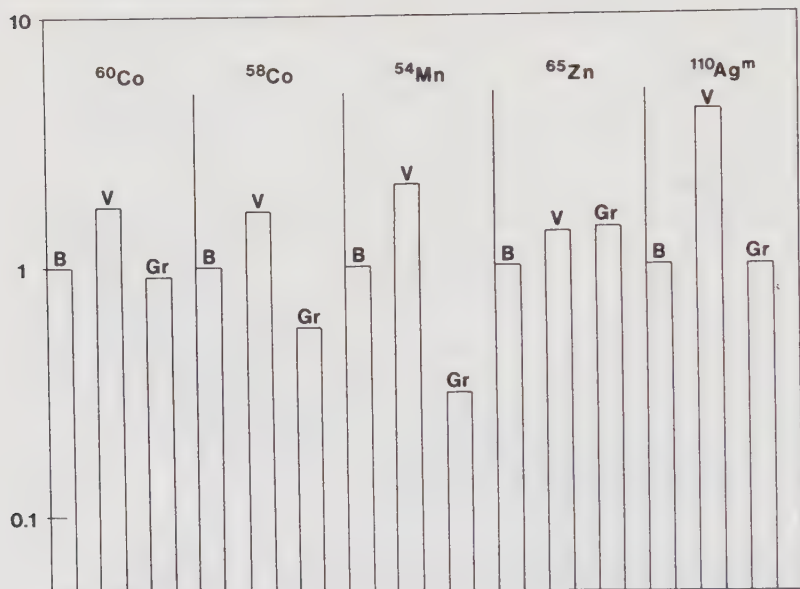


Figure 11. Activity concentrations, relative to *I. baltica*,
for activation products in *I. viridis* and *I. granulosa*.

Fucus v. in December 1978 at various distances from Barsebäck. The activity concentration in I. baltica well reflects the concentration in Fucus v. presumably due to the fact that Idothea is grazing on the seaweed plants. The ratio between the ^{60}Co activity concentration in I. baltica and Fucus v. was 0.16 ± 0.04 at that time. Considerably higher ratios, 0.93 ± 0.10 , are found in summer. This is illustrated in figure 10. The explanation for this seasonal variation may be the decreased metabolic activity of Idothea during the winter period.

In 1979 a very large sample of Idothea was taken at a location 1.2 km north of the discharge pipe. Over 3000 animals were separated giving separate samples of I. baltica, I. viridis and I. granulosa. Due to the large samples and the proximity to the power plant, an analysis of other radionuclides could be made. The result from this analysis is shown in figure 11, where the activity concentration is given as relative to I. baltica. It is evident that I. viridis, for all radionuclides, shows a higher uptake than I. baltica. The opposite result applies to I. granulosa. It is a well known fact (7) that these three species of the Idothea family behave differently. For instance, I. viridis can usually be found in the most still waters near the shore and I. granulosa in more turbulent areas. Idothea baltica are present in areas "in between". This difference can not explain the differences shown in figure 11, but it still indicates that other differences could exist, for instance with regard to their feeding-habits. This will hopefully be subject to further studies.

C. Comparative studies of activity concentrations in different algae and crustaceans.

The activity concentration in some algae and crustaceans relative to that of Fucus is given in figure 12. All concentrations are given on dry weight basis. It is interesting to note the differences in activity concentration in I. baltica and Gammarus living side by side in Fucus plants. For the cobalt isotopes the concentration in I. baltica is a factor of 4 higher than in Gammarus.

Activity concentration (d.wt.) relative to Fucus

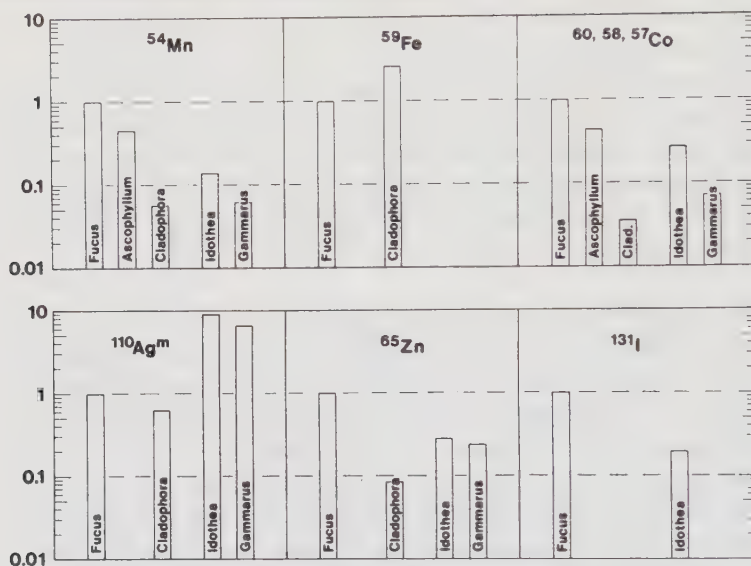


Figure 12. Relative content, per dry weight unit, in different algae and crustaceans of radionuclides released from Barsebäck in August-November 1979. Dry/wet weight ratios: Fucus-0.2, Ascophyllum-0.2, Cladophora-0.05, Idothea-0.19 and Gammarus-0.18.

This may indicate a difference in their feeding-habits. The highest activity concentration of ^{54}Mn , $^{60,58,57}\text{Co}$, ^{65}Zn and ^{131}I are found in Fucus while crustaceans for these nuclides show an activity concentration of 4-40% compared to Fucus. For $^{110}\text{Ag}^m$, the activity concentration in Idothea and Gammarus is a factor of 9 and 6 respectively, higher than in the Fucus plants in which they live. This finding indicates an active uptake of silver in these crustaceans. Because Idothea and Gammarus are eaten by various fish species, these findings may be of considerable importance for the transport of radioactive substances to man.

D. Studies using Mytilus edulis.

A limited number of samples of the common blue Mussel, Mytilus edulis were taken at some of the Fucus fields. The separate analysis of the shell and the muscle (body) of the mussle gave the activity ratios muscle/shell shown in table I (based on dry weight). (Dry weight/wet weight ratio for Mytilus (muscle) is ≈ 0.2).

Table I

Activity ratio Muscle/shell

^{60}Co	^{58}Co	^{54}Mn	^{65}Zn	$^{110}\text{Ag}^m$	^{40}K
2.4 ± 0.1	2.8 ± 0.7	0.82 ± 0.30	12.9 ± 2.9	1.9 ± 2.0	21 ± 6

The differences between cobalt, manganese and zinc are in agreement with those found by Dahlgaard (8). The great accumulation of ^{65}Zn in the soft parts should be stressed because of the radio-logical importance of the mussel as a part of important food-chains which finally lead to man.

In table II, a comparison between Mytilus edulis and Fucus is made. The ratio of activity concentrations on dry weight basis are given.

Table II

Activity ratio Mytilus (muscle)/Fucus

^{60}Co	^{58}Co	^{54}Mn	^{65}Zn	^{40}K
0.08 ± 0.01	0.07 ± 0.02	0.05 ± 0.01	0.18 ± 0.02	0.55 ± 0.05

The results clearly indicate that Fucus is superior to Mytilus for the purpose of mapping the geographical spreading of activation products.

E. ^{60}Co in water and sediment

The ^{60}Co activity concentration in unfiltered seawater 2.9 km NNW of the power plant was determined to 5.4 mBq/l (1978-06-15), 4.1 and 5.1 mBq/l (1978-09-22) and 1.3 mBq/l (1979-04-11). Our results, indicate that the concentration factor for Fucus (dry weight) is as high as $(2\pm 1) \cdot 10^5$ corresponding to a wet weight concentration factor of $(4\pm 2) \cdot 10^4$. This value is considerably higher than those computed by Fukai and Murray (9) or measured in aquariums by Nakahara et al. (3). It is interesting to note that the concentration factors for actinides in this area have also been found to be unusually high (10). Measurable concentrations of ^{60}Co have been found in sediments from sampling stations NW of the power plant 1.7-2.5 km off-shore. The total area content of ^{60}Co was found to be 190 Bq/m² at 4.1 km from the plant, 76 Bq/m² at 6.4 and 560 Bq/m² at 11 km. Around 70% of the ^{60}Co activity was found in the upper 25 mm of the sediment layer and the rest in the next 25 mm.

SUMMARY AND CONCLUSION

The macroalgae Fucus vesiculosus has, in comparison with other common algae, proven to be a very suitable and sensitive bioindicator for mapping the release of activation and fission products from a nuclear power plant into the marine environment. The activity concentration in Fucus along the coast can be expressed using a power function (cf. eq. 2). It has been shown that the constant α well reflects the discharge rate pattern, indicating that Fucus is a suitable indicator with regard to the activity concentration in the surrounding water. The small variations of the constant β with time and between most radionuclides indicate that the spreading pattern is extremely constant in time, which makes the Öresund area suitable for radioecological studies.

Studies of the crustaceans Idothea have shown that the activity concentration of ^{60}Co varies between winter and summer. Within the Idothea family, the ability to concentrate activation products differs remarkably between species. The mussel Mytilus edulis generally show lower activity concentration values than Fucus. The high accumulation

of ^{65}Zn in the soft parts should be noted. Because Idothea as well as Mytilus are eaten by various fish species these findings may be of importance for the transport of radioactive substances to man.

ACKNOWLEDGEMENT

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MATHEMATICAL MODELS FOR SIMULATION OF PROCESSES OF RADIOECOLOGICAL
AND BIOMEDICAL TRANSPORT OF RADIONUCLIDES

Part III. Application on ^{131}I in the food-chain
grass-cow-milk-man following a BWR 1
accident.

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INTRODUCTION

Any operation of nuclear fission reactors will inevitably give rise to releases of radioactive nuclides to the environment. These releases will result in increased individual and collective dose equivalents especially to people living in the vicinity of the reactor. However, mechanisms exist, which through mechanical and ecological transport will increase the dose levels for people living at greater distances from the plant. Some of these will be discussed in detail in later sections.

The impact of such releases on individuals and populations can roughly be divided into three components:

- 1) External irradiation from the atmosphere into which radionuclides are released
- 2) External irradiation from the ground and other structures onto which radionuclides are deposited from the atmosphere
- 3) Internal irradiation following inhalation of radionuclides and/or intake of food from the terrestrial or marine environment containing radioactive nuclides.

Briefly one could say that the normal operation of a nuclear reactor gives rise to releases which result in dose increases which are totally negligible compared to the dose contribution from other man-made sources of radiation used in daily life. However, any minor malfunction or incident in the operation of the reactor could result in an increased dose level, above the maximum permissible dose equivalent, to people living nearby the plant. In order to establish the maximum levels of the activity releases during operation giving rise to dose equivalents well below accepted limits, the need for knowledge regarding the behaviour of radionuclides in the environment is evident.

Studies giving such information have been carried out during the last 30 years. The source of the radionuclides has above all been fallout from nuclear weapons tests, but also specific radionuclides have been used in laboratory and field experiments. The techniques for measurement have gradually been improved, resulting in possibilities to detect still lower activity values. Thus, reprocessing plants and later nuclear reactors as sources for various radionuclides have more and more been used giving

valuable information also of the characteristics of the releases from a specific source at a specific site.

The number of radionuclides studied is, in spite of the 30 year research period, quite limited. However, for cesium, strontium, plutonium and americium isotopes, detailed information can be found in the literature. The sources for these studies are nuclear fallout and discharges from re-processing plants. For some isotopes, only laboratory and field experiments give information of their behaviour, and often in very limited parts of the biosphere. The application of such data to a different source should thus be made with great care and caution. One must for example consider specific properties of the source and its surroundings that could affect the validity of the data given by experiments under controlled (man-made) conditions.

This situation applies to the radioactive nuclei of iodine, where very much experimental work indeed has been carried out. A great deal of these studies have been focussed on the uptake and secretion of ^{131}I into the milk of grazing cattle under conditions simulating environmental contamination of pasture resulting from fission bombs and reactor discharges.

The purpose of this work is to study the transfer of ^{131}I in the pathway pasture-cow-milk-man and the resulting dose equivalents (and dose commitments) to a population within 120 km from a nuclear power plant in southern Sweden.

In order to carry out this study, one then has to penetrate important properties and behaviour of every single link in this chain. The chain is in the following paragraphs split into the following sections:

- I) The reactor core
- II) The spreading and deposition pattern
- III) The metabolism of iodine in the dairy cow
- IV) Factors affecting the individual dose equivalents and collective dose equivalents

The following text will treat these sections in the order I - IV above.

I. The reactor core as a source for ^{131}I .

The nuclear power reactors of interest in this study are found at the Barsebäck nuclear power plant, located on the Swedish west coast at the Öresund, 20 km east of Copenhagen. The power plant consists of two reactors, each of 1700 MW (thermal) effect.

The ^{131}I -activity in the reactor core after $2 \cdot 10^6$ MWD burnup is approximately 1700 PBq (46 MCi). The fraction of this activity that is released in case of an accident naturally depends on the nature of the accident as well as the function (or malfunction) of the systems designed to prevent overheating of the core. Different types (or classes) of reactor accidents have been defined and were extensively studied in the WASH - 1400 report.

The amount of iodine that is released strongly depends on the thermal rest-effect and/or the amount of water remaining in the reactor vessel or -containment into which the iodine in inorganic form can be dissolved. The accident-categories given by WASH - 1400 implies that between 1% and 90% of the iodine is released in inorganic form. The contribution from organic-bound iodine is negligible.

In this study a release of 100% of the iodine present in one of the cores in connection with a BWR 1 accident is assumed. Lower figures could be applied by simply reducing all the figures in the final results (dose figures) by the desired fraction. The activity is supposed to be released during a time which is short compared to the physical half-life of ^{131}I (8.05 d).

II. The spreading and deposition pattern.

Due to the relatively high thermal rest effects in a reactor during a number of hours following shutdown, the activity released will be carried upwards to a considerable height. This height which is dependent also on the wind-speed, can be from approximately 100 to 1000 m above ground. The atmosphere spreading can be calculated in a number of ways. One of the most common methods is to use Brigg's spreading model (Gifford, 1976). This model utilizes the Pasquill weather classification of atmospheric stability, divided into six classes, A - F. The "A" class

represents the most unstable type of atmosphere with the most variable wind direction and class "F" represents the most stable conditions. Table I gives some properties characterizing the different classifications A-F at the Atlantic coast (WASH - 1400)

Table I

	A	B	C	D	E	F	Total
Wind speed (m/s)	2-5	3-6	2-6	2-6	1-3	1-3	1-5
Time without rain %	4	5	2	36	30	16	95
Time with rain %	0	0	0	2	2	0.9	5
Std dev in							
horizontal wind	25°	20°	15°	10°	5°	2.5°	-
direction							

Of the six classes given above, Pasquill D is the most frequent at the Barsebäck site. In table II statistics for Malmö (20 km south of Barsebäck) for the different types can be seen.

Table II

Wind direction (from)	Percent of time with weather of type			
	B	D	E	F
S	5	74	17	4
W	3	81	13	3
N	6	53	30	11
E	3	62	28	8

Because of its high frequency, Pasquill D has been used in the study together with a wind speed of 6 m/s which is the most common wind speed in this area.

Weather of class Pasquill F is relatively seldom found in these areas. Yet it has been included in the study because the presence of this type results in the most distant spreading of radionuclides due to the stable wind direction and air inversion. The wind speed is here assumed to be 3 m/s.

The time-integrated activity concentration in air near the ground at the point (x, y, z) can be expressed, using a two-dimensional normal distribution, as:

$$\bar{C}(x,y,z,H,S,v) = \frac{1}{2\pi \sigma_y(x,S) \cdot \sigma_z(x,S)} \cdot \int_0^t \frac{dA(t)}{dt} \cdot e^{-\lambda \cdot \frac{x}{v}} dt \cdot e^{-\frac{y^2}{2\sigma_y^2(x,S)}} \cdot \left[e^{-\frac{x^2}{2\sigma_z^2(x,S)}} + e^{-\frac{(z+H)^2}{2\sigma_z^2(x,S)}} \right] \quad (1)$$

where

H = the effective release height (m)

S = atmospheric stability class

v = wind velocity (m/s)

$\sigma_y(x,S)$ = horizontal dispersion parameter (m)

$\sigma_z(x,S)$ = vertical dispersion parameter (m)

$\frac{dA(t)}{dt}$ = activity release rate (Bq/s or Ci/s)

λ = physical decay rate constant (s^{-1})

The dispersion parameters are thus dependent both on the stability class and the distance from the release point. As an example, the values of σ_y and σ_z for class Pasquill F are given in table III.

Table III

Distance (km)	σ_y (m)	σ_z (m)
1	35	13
2	65	22
5	150	34
10	270	50
20	500	60
50	1120	80

The theories for geographical spreading have been studied within a distance of 120 km from the power plant. For this purpose the area has been divided into small area elements consisting of circle segments.

Figure 1 illustrates the principle of this system.



The circle is divided into 36 sectors, each of an opening angle of 10° . The distances from the plant are taken in fifteen distance intervals, ten of six kilometers each up to 60 km from the plant and five of twelve kilometers each from 60 to 120 kilometers from the plant. The notation for a specific sector is in the following given as a matrix index (i,j) corresponding to angle and distance (θ,D) , where

- 1) the angle θ (degrees) refers to the middle of the sector, and is expressed as $\theta = i \cdot 10^{-5}$

2) the distance D (km) refers to the middle of the distance interval and is expressed as

$$D = \begin{cases} 6 \cdot j - 3 & \text{for } 1 < j < 10 \\ 12 \cdot j - 66 & \text{for } 11 < j < 15 \end{cases}$$

This will require a matrix consisting of 540 elements for storage of data in the following analysis.

The horizontal dispersion will affect the spreading pattern so that not only sectors directly in the wind direction will be subject to deposition of radionuclides. Pasquill A will give rise to the greatest horizontal spreading and Pasquill F the smallest. Table IV gives the deposition levels, normalized to the levels in sectors in the wind direction, for adjacent sectors.

Table IV

Distance (km)	Pasquill D		Pasquill F
	$\Theta \pm 10^0 (i \pm 1)$	$\Theta \pm 20^0 (i \pm 2)$	$\Theta \pm 10^0 (i \pm 1)$
0 - 6	0.30	$3.6 \cdot 10^{-4}$	$2.9 \cdot 10^{-2}$
6 - 12	0.24	$2.3 \cdot 10^{-5}$	$9.0 \cdot 10^{-3}$
12 - 18	0.18	$9.7 \cdot 10^{-7}$	$2.3 \cdot 10^{-3}$
18 - 24	0.15	$1.2 \cdot 10^{-7}$	$9.0 \cdot 10^{-4}$
24 - 30	0.11	$4.7 \cdot 10^{-9}$	$2.2 \cdot 10^{-4}$
30 - 36	0.07	$6.7 \cdot 10^{-11}$	$3.2 \cdot 10^{-5}$
36 - 42	$5.7 \cdot 10^{-2}$	$7.4 \cdot 10^{-12}$	$1.2 \cdot 10^{-5}$
42 - 48	$3.6 \cdot 10^{-2}$	$1.0 \cdot 10^{-13}$	$1.9 \cdot 10^{-6}$
48 - 54	$2.9 \cdot 10^{-2}$	$1.2 \cdot 10^{-14}$	$7.1 \cdot 10^{-7}$
54 - 60	$1.8 \cdot 10^{-2}$	$1.5 \cdot 10^{-16}$	$1.1 \cdot 10^{-7}$
60 - 72	$1.1 \cdot 10^{-2}$	$2.0 \cdot 10^{-18}$	$1.6 \cdot 10^{-8}$
72 - 84	$5.5 \cdot 10^{-5}$	$3.0 \cdot 10^{-21}$	$9.1 \cdot 10^{-10}$
84 - 96	$2.7 \cdot 10^{-3}$	$4.5 \cdot 10^{-24}$	$5.1 \cdot 10^{-11}$
96 - 108	$1.3 \cdot 10^{-3}$	$6.8 \cdot 10^{-27}$	$3.0 \cdot 10^{-12}$
108 - 120	$6.5 \cdot 10^{-4}$	$1.0 \cdot 10^{-27}$	$1.7 \cdot 10^{-13}$

The deposition on the ground of radionuclides can take place under dry or rain (snow) weather conditions. For the dry deposition case, the initial activity per square meter $Ag(0)$ can be expressed by using the time-integrated activity concentration in air near the ground $\bar{C}(x, y, z, H, S, v)$ (cf equation 1) as

$$Ag(0) = \alpha \cdot \bar{C} \quad (2)$$

where α is the deposition velocity (m/s). The value used in this study for this coefficient is $3 \cdot 10^{-3}$ m/s. (The WASH - 1400 report gives an interval of 0.1 - 0.001 m/s). Deposition during precipitation is assumed to occur in a way so that a certain fraction of the plume content is washed out per unit time. This fraction is in this study taken as $10^{-4} (s^{-1})$. (The WASH - 1400 report gives an interval of $10^{-2} - 10^{-5} (s^{-1})$).

In figures 2 and 3 the area content of ^{131}I (here expressed as $mCi \cdot m^{-2}$) as a function of distance after a release of 46 MCi ^{131}I from a BWR 1 accident is given. No correction for decay during time from release to deposition has been made, since the maximum time of transport is eleven hours (to 120 km). Figure 2 applies to Pasquill D and figure 3 to Pasquill F condition. The great difference between wet and dry deposition is especially observable for Pasquill F at short distances. The right hand ordinate is treated in the following section (III).

We are now in position to calculate the area content in all sector (matrix) elements. The deposition time is in the following assumed to be very short compared to the half-life of ^{131}I .

For a certain weather situation the area contents, at distances corresponding to the middle of the distance intervals in the circle segments, are taken from figure 2 or 3. If the wind direction is in angle θ , the area contents mentioned above are stored in the elements of column nr i in the data matrix, where $i = (\theta + 5)/10$. The angle θ must be expressed as 5° , 15° , 25° etc. The contribution from the horizontal dispersion is calculated with the aid of table IV above, and is distributed in the matrix in row $i \pm 1$ and, for Pasquill D also in row $i \pm 2$.

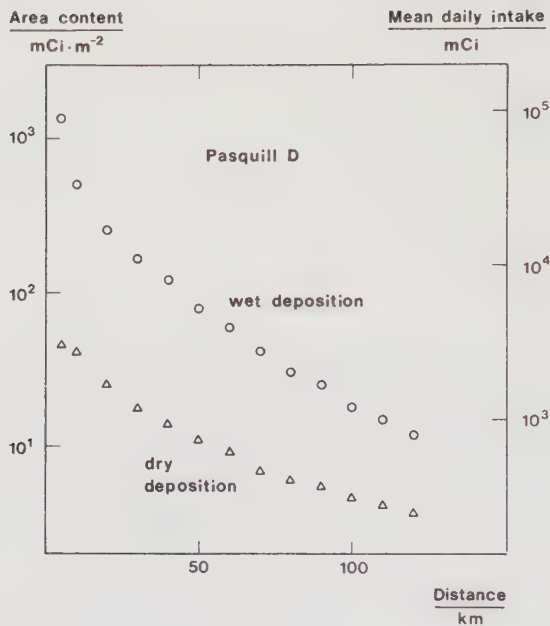


Figure 2

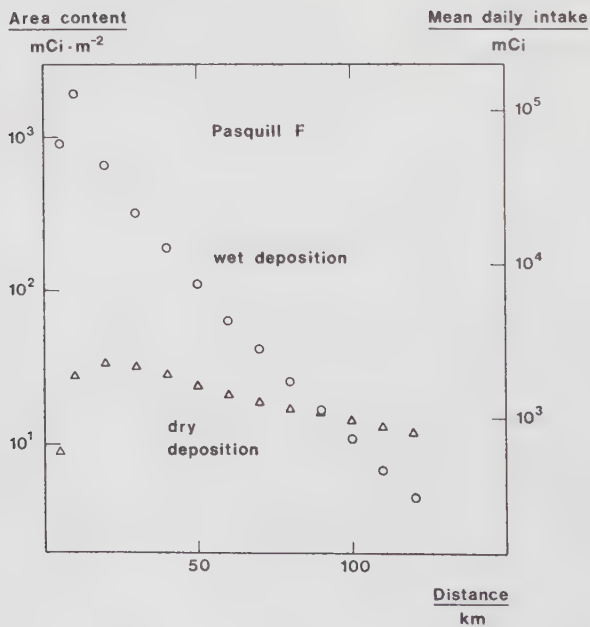


Figure 3

The initial area content ($\text{Bq}\cdot\text{m}^{-2}$ or $\text{Ci}\cdot\text{m}^{-2}$) in circle segment (θ , D), or matrix elements (i , j) is in the following analysis given as

$$\text{Ag}(\theta, D) \text{ or } \text{Ag}(i, j)$$

The effective half-life of ^{131}I on the ground is, of course, shorter than the physical half-life of ^{131}I (8.05d). Experiments by Sasser and Hawley (1966) imply that the half-time is 5.5 ± 0.8 days. Other workers in this field have not taken a shorter half-time on ground into account. As the purpose of this work is to estimate radiological rather than chemical consequences, this faster reduction of ^{131}I available to grazing animals has not been considered.

III. Metabolism of iodine in the dairy cow.

This section treats a subject that since long intensely has been studied and reported in a great number of publications. Of the most renowned persons in this field, Comar, Lengemann and Garner should be mentioned.

An excellent summary of the work performed in this field is found in an article by Booth, Burke and Kaye (Oak Ridge). The compartment model in this work utilizes much data from the Oak Ridge Summary. The model has, however, been modified and adopted to conditions representative for the Swedish cow and its diet (Ekman and Jones, 1980).

The conditions for which compartment analysis in a transport system is applicable has been discussed in part I of this article series and is assumed to be valid in this analysis.

In figure 4 the compartment system is presented. It consists of 16 compartments with two input and three exit compartments. All of these are not used in the iodine model; transport to and from meat, liver and skeleton is not considered. The reason for their presence is that the model with few changes could be applied also for metabolism of cesium and strontium.

The 17 transport coefficients between the compartments follows the definition given in part I (numerical methods) and are listed in table V below.

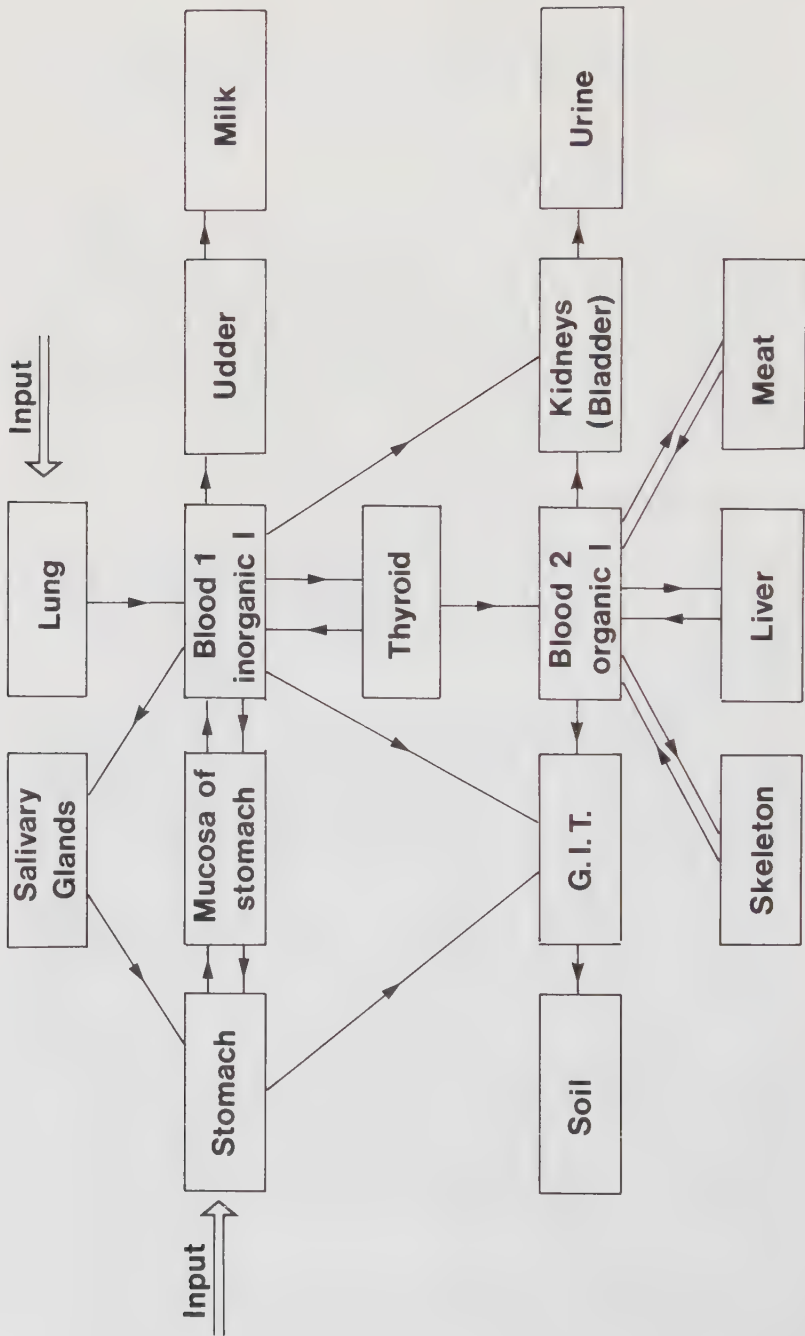


Table V

Transport coefficients (P) in the cow model (unit:d⁻¹)

From	To	P(d ⁻¹)
Stomach (mucosa)	Blood 1	0.950
Stomach	GIT	0.432
Blood 1	Thyroid	0.280
Blood 1	Udder	0.200
Blood 1	GIT	0.518
Blood 1	Kidneys	0.371
Blood 1	Salivary glands	0.050
Blood 1	Mucosa of stomach	0.050
Thyroid	Blood 1	0.066
Thyroid	Blood 2	0.043
Udder	Milk	2.00
GIT	Soil	1.00
Blood 2	GIT	1.00
Blood 2	Kidneys	0.033
Kidneys	Urine	3.00
Salivary glands	Stomach	0.900
Mucosa of stomach	Stomach	0.900

In case of input via lung, all the activity immediately is transferred to the blood 1 compartment. The input is thus treated as if the injection was directly into the inorganic blood compartment.

The blood is, as can be seen, divided into two compartments containing inorganic and organically bound iodine, respectively. From table V it can be seen that excretion into milk of organically bound iodine is omitted. This simplification relies on the fact that, according to Lengemann and Comar (1963) only about 5% of the iodine in milk is present in organic form. The percentage of the excreted iodine in organic form can then possibly not be greater than 5%.

The compartment analysis for calculation of the transport of ¹³¹I in the cow consists of 17 coupled linear differential equations. The integration of the system is carried out using the BIOWAY computer program described in part II (A computer program) of this article series. The methods of solution is either the Kutta-Merson method or the reduced

serial expansion method described and discussed in part I (numerical methods).

SENSITIVITY ANALYSIS AND RESULTS

From figure 4 and table V it is evident that factors which affect the amount of iodine that is transferred to the udder only are the competing ways of transport out of the blood 1 compartment. Of the five possible paths it is only three which are of enough magnitude to affect the udder content. These are transport to kidneys, GIT and thyroid. Reduction of any of the transport coefficients to these compartments automatically results in an increased transport to the udder, and vice versa. In the sensitivity analysis, these parameters have been assumed (Ekman & Jones, 1980) to vary between the limits given in table VI below.

Table VI

From	To	Interval (d^{-1})	Nominal value (d^{-1})
Blood 1	Kidneys	0.20 - 0.67	0.371
Blood 1	GIT	0.20 - 1.50	0.518
Blood 1	Thyroid	0.05 - 0.60	0.280

The calculations are made using an input into the stomach of 37 GBq (1 Ci) during one day, that is, a pulse during only this single day. The results are presented in the figures 5, 6, 7 and 8.

Figures 5, 6 and 7 shows the effects of different excretion rates to kidneys, faeces and thyroid, respectively. In figure 8 the extremes have been combined, giving the highest and lowest possible iodine content in the milk. It should be pointed out that these figures refer to the accumulated activity in the milk as a function of time; not the activity excreted daily into the milk.

From figure 8 it is found that a peak activity of between 3.2% and 11% of the activity taken in by the cow is excreted into the milk, in which a maximum activity is reached after 4-6 days. The "normal" value is approximately 6%. These values are in good agreement with those reported from field experiments by Sasser and Hawley. They found a median

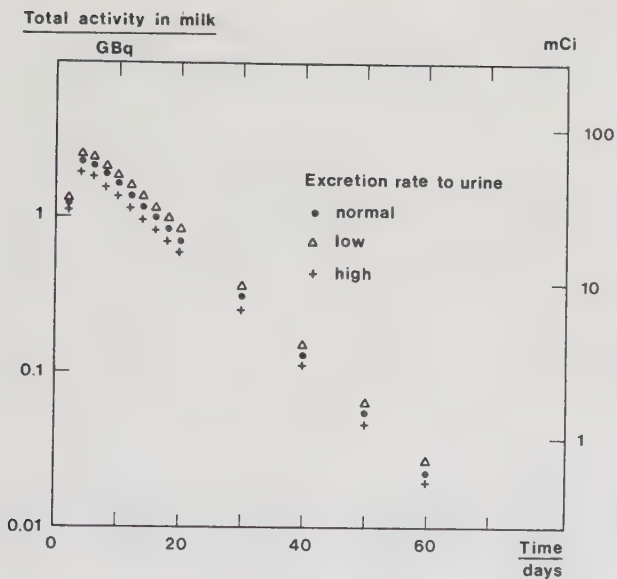


Figure 5

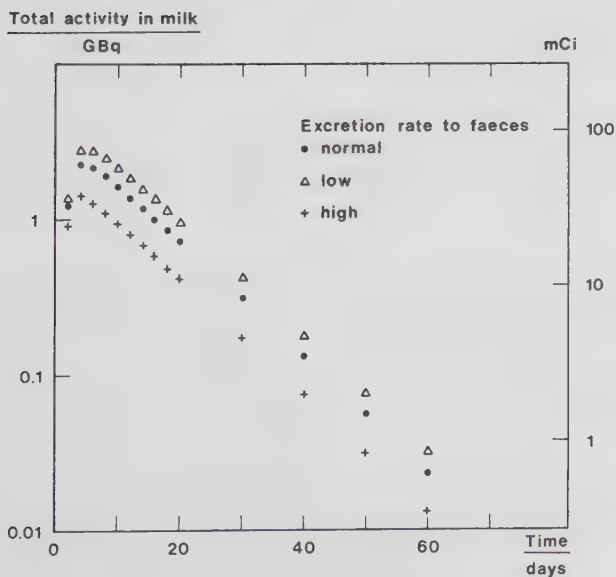


Figure 6

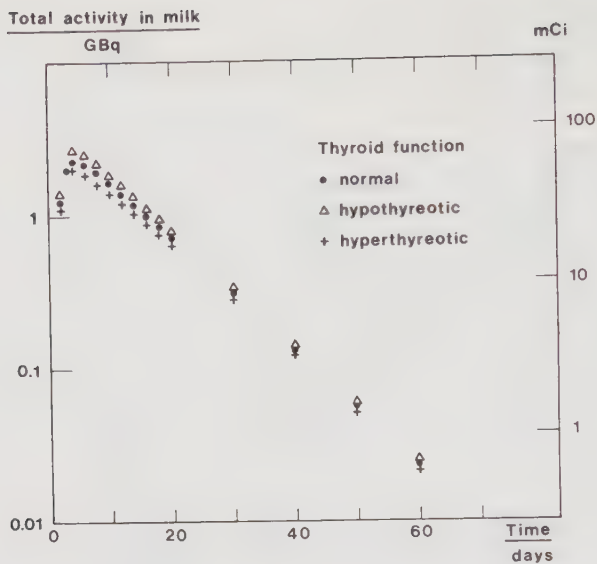


Figure 7

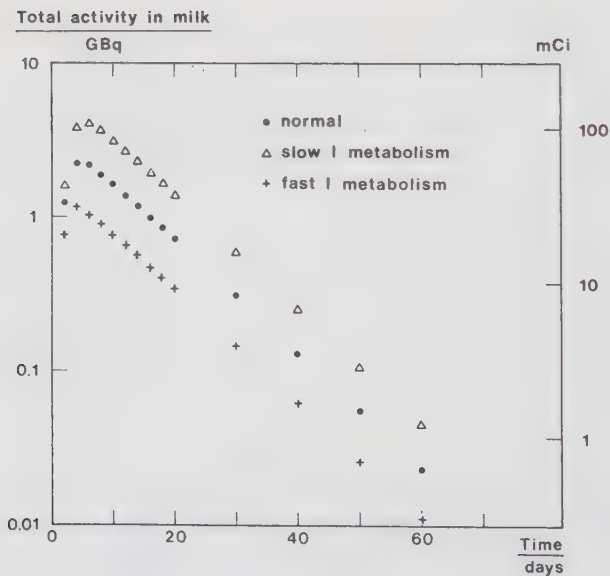


Figure 8

value of 6% with maximum and minimum values of 29% and 4%, respectively. Their figures are based on activity totally recovered in milk over a 14-days period, but due to the short half-time of iodine in milk after exposure (0.7 days) the values are almost identical to peak values. The values also agree with those of Potter et al. (16) and the CERT tests (2) which give maximum and minimum values of 12% and 4.5% respectively, assuming a milk production of 15 liters per day. Assuming this milk production, the data given by Garner and Jones (1960) and Garner (1967) are minimum, mean and maximum peak values of 3.4%, 8.9% and 18.4%, respectively. Garner, Sanson and Jones (1960) present results on transfer of ^{131}I as iodide and iodine. Their mean recovery was 6.7% and 7.3% at a milk yield of about 10 liters per day.

The faeces and urine data calculated with the model used in this work also agree well with those given by other workers. The model can therefore be expected to describe the metabolic conditions for iodine with an acceptable precision for the application outlined in this work.

From the literature, for example from data by Lengemann and Comar (1964), it is evident that the amount of iodine that is excreted into milk is closely related to the volume of milk that is produced. The milk volume in itself is not believed to be totally responsible for the amount of activity that is excreted. These workers found that individual variations of concentrating mechanisms are responsible for variations in the milk/plasma ratio. Still, the relation between activity and volume remains. This, of course, results in a nearly constant activity per unit volume in the milk.

The variation in milk production over the year is closely related to the time of birth of the calves. For economical reasons, it is in Sweden tried to concentrate the births to the autumn period. The milk production is at this time higher for the individual cow. A somewhat lower milk production is found in the beginning of the grazing period (May) before the cows have gotten used to their new feeding-conditions. In a large population of cows, these variations are small (Jones, 1980) and as the human consumption in the analysis below is based on volume rather than activity consumption the effect of these variations are negligible. A daily constant milk production rate is thus assumed.

The results used in the following analysis for calculation of absorbed doses to the population are, of course, not the results from the sensitivity analysis. For this application, the output rate into milk expressed as, normalized to the initial activity input rate, a function of the activity input rate with time into the stomach (lung) of the cow, is used. This quantity is in the calculations found as

$$\frac{\epsilon_m(t)}{\frac{dA_0}{dt}} = \frac{A_{udder}(t) \cdot P_{udder \rightarrow milk}}{\frac{dA_0}{dt}} = f\left(\frac{\frac{dA(t)}{dt}}{\frac{dA_0}{dt}}\right) \quad (3)$$

where

$A_{udder}(t)$ = activity at time t in udder

$P_{udder \rightarrow milk}$ = transport coefficient from udder to milk

$\frac{dA(t)}{dt}$ = activity input rate into stomach (lung)

This quantity is thus in the following analysis given as $\epsilon_m(t)$.
($\epsilon_{m, lung}(t)$).

IV. Factors affecting the individual and collective dose equivalents (and commitments). Dose calculations.

The impact of a release of ^{131}I following a BWR 1 accident has been analyzed for the following situations:

- a) Accident in the period October-April
- b) Accident in the period May-September
- c) Pasquill D, dry deposition
- d) Pasquill D, wet deposition
- e) Pasquill F, dry deposition
- f) Pasquill F, wet deposition

These result in eight different situations for which results will be presented.

A basic condition for the validity of the calculations is the fact that all of the activity excreted into milk that is produced in the 120 km zone is consumed with known time delays between production and consumption. In other words, this requires that a known fraction of the milk produced is delivered to the dairies. Knowledge of the fractions of milk that is used for different products (cream, cheese etc) is also required. The fraction of milk that is used for direct consumption at the farm is set to zero, but is still considered for the calculation of the maximum individual absorbed doses.

Prior to the presentation of the analysis it must strongly be stressed that all dose figures only apply to the intake of ^{131}I present in dairy products.

The calculations will be presented for one circle segment (θ, D). The index for this segment is given as the letter i . No specific symbols to distinguish between the situations a) - f) above are used.

ABSORBED DOSE CALCULATIONS

The total number of people present in the sector i is T_i . This population is divided into three groups: infants 0-5 years of age, children 5 - 15 years, and adults >15 years.

These groups are called S_i , B_i and V_i , respectively. Their fractions of T_i are assumed to be the same in all sectors as the national Swedish population statistics from 1975 (FOB, 1975). They are:

$$\begin{aligned} S_i &= 0.065 \cdot T_i = f_s \cdot T_i \\ B_i &= 0.14 \cdot T_i = f_b \cdot T_i \\ V_i &= 0.795 \cdot T_i = f_v \cdot T_i \end{aligned} \quad (4)$$

The average consumption rate of milk by these groups are given as F_s , F_b and F_v .

If a total activity A_i is available for consumption by T_i individuals, the total activity that is separately consumed by the respective groups will be A_{is} , A_{ib} , A_{iv} .

$$\begin{aligned}
 \text{Infants: } A_{is} &= A_i \cdot \frac{S_i \cdot F_s}{S_i \cdot F_s + B_i \cdot F_b + V_i \cdot F_v} = \frac{A_i \cdot f_s \cdot F_s}{f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v} \\
 \text{Children: } A_{ib} &= \frac{A_i \cdot f_b \cdot F_b}{f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v} \quad (5) \\
 \text{Adults: } A_{iv} &= \frac{A_i \cdot f_v \cdot F_v}{f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v}
 \end{aligned}$$

The uptake of this activity results in a distribution pattern within the body that is similar to what is described in the cow model in section III. The most important doses to be considered are the thyroid doses and weighted whole-body doses as defined by the ICRP (ICRP publication 26). The conversion factors (dose/activity) for the three groups substantially differ. Table VII gives some data on the thyroid gland and its uptake and retention of iodine. Data are from Soldat et al. (1974).

Table VII

Age group	Uptake %	Biological $T_{1/2}$ (d)	mass(g)
0 - 5	50	15	2
5 - 15	40	30	10
> 15	30	100	20

The figures below are given in units referring to the old activity unit Ci. This is done only in order to simplify comparison with literature data.

For adults, the dose conversion factor for the thyroid is (SFR,1979)

$$C_{v,t} = 20 \text{ kGy/Ci}$$

The weighted whole body dose is (SFR, 1979)

$$C_{v,w} = 0.6 \text{ kGy/Ci}$$

In the case of infants, the dose conversion factor for the thyroid $C_{s,t}$ is different due to

- a) the lower weight (0.1 of the adult)
- b) the more rapid excretion
- c) a higher uptake

The effective half-life in the thyroid is 5.1 days. This gives a different number of decays in the thyroid compared to adults. The reduction factor is

$$\frac{\int_0^{\infty} e^{-\lambda_s \cdot t} dt}{\int_0^{\infty} e^{-\lambda_v \cdot t} dt} = \frac{\lambda_v}{\lambda_s} = 0.69$$

The results of a), b) and c) above is that the thyroid absorbed dose is 11.5 times higher for infants, that is 230 kGy/Ci. The weighted whole-body dose is predominantly dependent on the thyroid dose and is approximately 7 kGy/Ci.

For the age-group 5 - 15 years, the figures for thyroid and weighted whole-body doses would be 45 kGy/Ci and 1.4 kGy/Ci, respectively. Table VIII below gives the dose-conversion factors for some different organs for ^{131}I expressed in kGy/Ci.

Table VIII

Age-group	Thyroid	Gonads	Bone-marrow	Liver	Whole body*
Infants	230	} 0.03	} 0.09	} 0.15	7
Children	45				1.4
Adults	20				0.6

*weighted according to ICRP 26

It is now possible to calculate the individual absorbed doses to a specific organ (0) of a person in one of the three age-categories. Using the expression (5), the doses in sector i is, for

$$\begin{aligned}
 \text{Infants } D_{is,o} &= \frac{A_{is}}{S_i} \cdot C_{s,o} = \frac{A_i \cdot f_s \cdot F_s \cdot C_{s,o}}{(f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v) \cdot S_i} \\
 \text{Children } D_{ib,o} &= \frac{A_{ib}}{B_i} \cdot C_{b,o} = \frac{A_i \cdot f_b \cdot F_b \cdot C_{b,o}}{(f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v) \cdot B_i} \\
 \text{Adults } D_{iv,o} &= \frac{A_{iv}}{V_i} \cdot C_{v,o} = \frac{A_i \cdot f_v \cdot F_v \cdot C_{v,o}}{(f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v) \cdot V_i}
 \end{aligned} \tag{6}$$

The collective doses to a specific organ (o), here called CD_i are then found from the expression (6) as

$$CD_{i,o} = D_i \cdot T_i \tag{7}$$

or for the individual groups

$$\begin{aligned}
 \text{Infants } CD_{is,o} &= \frac{A_{is}}{S_i} \cdot C_{s,o} \cdot S_i = A_{is} \cdot C_{s,o} \\
 \text{Children } CD_{ib,o} &= \frac{A_{ib}}{B_i} \cdot C_{b,o} \cdot B_i = A_{ib} \cdot C_{b,o} \\
 \text{Adults } CD_{iv,o} &= \frac{A_{iv}}{V_i} \cdot C_{v,o} \cdot V_i = A_{iv} \cdot C_{v,o}
 \end{aligned} \tag{8}$$

It should be noted as seen from (8), that the collective dose not is dependent on the number of people that is exposed to the radioactive material.

One parameter remains to be accounted for, namely the milk consumption. In Sweden, the average consumption (1975) of milk and cream is 0.48 liters per person and day. The infants is in this study assumed to consume 0.5 l/day, the children and adults 1.0 and 0.5 l/day, respectively. This would give a mean value of 0.57 liters per person and day which is 18% higher. However, the milk consumption in Sweden tends to increase with about 2% per year, thus making this higher value more realistic.

In table IX, all values for the three age-groups used in the collective dose calculations are summarized.

Table IX

	Infants (0-5 years)	Children (5-15 years)	Adults (> 15 years)
Percentage of population f(%)	6.5	14	79.5
Daily milk intake F(l/d)	0.5	1.0	0.5
Dose conversion factor for thyroid (kGy/Ci)	230	45	20
Dose conversion factor for whole body (ICRP 26)	7	1.4	0.6
Dose conversion factor for whole body (ICRP 2)	0.11	0.02	0.01

For comparison the data for whole body absorbed dose prior to the introduction of the philosophy given in ICRP 26 are given.

The collective dose to the whole population involved can now be calculated by summing up the system (6) all over the circle sectors (i). This, however, requires knowledge of the activity available for intake, A_i . This quantity is being described in the following sections.

Activity available for intake, A_i

The activity of ^{131}I available for intake, that is the activity present in milk for consumption produced in sector i, is affected by a number of parameters. Listed below are those which are included in the study. These are

- a) The number of cows present in sector i
- b) The fraction of cows that actually produces milk
- c) Time of year. This will affect the ratio of grazing/stabled cows.
- d) The area (m^2) that is grazed per day and inhalation rate that gives rise to intake of ^{131}I .
- e) The fraction of milk that is delivered to the dairies
- f) The fraction of the delivered milk that is used for the production of consumption milk (and cream)
- g) The excretion of ^{131}I into milk as a function of time after intake of ^{131}I by grazing or inhalation following a known function of time.

These items will now be treated one by one

- a) The number of cows present in sector i. Table X below gives the number of cows in sector i, that is in sector (θ, D)
for $\theta = 5^\circ, 15^\circ, 25^\circ, \dots$
and $D = 3, 9, 15, \dots, 57, 66, 78, \dots, 108$
Data from Wickman (1979). Note: No data on cows in Denmark are given.
The number of cows in sector i is hereafter called N_i .

- b) The fraction of cows that actually produces milk in southern Sweden is 0.27. This has already been included in table X.

- c) Time of year. The year is divided in two periods:
"Summer", ranging from May - September, and "Winter", ranging from October - April.
For the summer period the following data are applied:
The fraction of cows that never are outside the cow-house is 0.1.
A fraction of cows are fed with fresh forage just outside the cow-house. These cows are inside the building during the nights.
This fraction is 0.15.
The fraction of cows that are outdoors grazing day and night, which is 0.75.

The intake of ^{131}I for these fractions is assumed to be:

- 1) For the 0.1 fraction: intake by forage is the same as for grazing cows. This is probably an overestimation of the real intake due to the fact that the fresh forage taken in from the fields does not

TABLE X

AVSTAND(KM)	5	15	25	VINKEL(GRADER)			55	65	75	85
				35	45					
3	8.	16.	16.	16.	15.		14.	8.	0.	2.
9	19.	27.	38.	40.	38.		41.	33.	25.	29.
15	13.	54.	86.	71.	115.		68.	40.	39.	36.
21	134.	86.	83.	147.	115.		145.	181.	111.	137.
27	306.	144.	169.	210.	168.		274.	151.	133.	158.
33	82.	187.	283.	216.	333.		455.	307.	394.	356.
39	95.	206.	271.	253.	361.		241.	182.	287.	760.
45	252.	392.	381.	269.	293.		250.	233.	428.	1205.
51	353.	456.	363.	191.	309.		281.	375.	704.	1602.
57	509.	324.	354.	364.	274.		403.	613.	617.	1515.
66	1275.	583.	617.	872.	905.		953.	953.	1098.	1260.
78	2442.	2469.	683.	480.	1240.		1439.	1050.	780.	749.
90	1052.	1366.	437.	378.	940.		1357.	1469.	709.	9.
102	1291.	807.	387.	310.	392.		660.	692.	516.	0.
114	983.	574.	645.	402.	440.		485.	825.	425.	0.

AVSTAND(KM)	95	105	115	VINKEL(GRADER)			145	155	165	175
				125	135					
3	2.	3.	3.	0.	0.		0.	0.	0.	0.
9	23.	13.	27.	21.	0.		0.	0.	0.	0.
15	39.	11.	40.	47.	82.		39.	0.	0.	0.
21	188.	60.	68.	47.	53.		48.	4.	35.	96.
27	133.	77.	116.	131.	131.		59.	30.	16.	12.
33	273.	20.	180.	131.	110.		100.	121.	15.	0.
39	469.	44.	150.	120.	211.		117.	76.	20.	7.
45	709.	171.	198.	593.	169.		90.	68.	2.	0.
51	1182.	505.	241.	702.	183.		70.	19.	0.	0.
57	1778.	1536.	446.	468.	85.		0.	0.	0.	0.
66	1936.	3122.	1526.	160.	0.		0.	0.	0.	0.
78	1149.	2136.	1531.	54.	0.		0.	0.	0.	0.
90	76.	1048.	575.	0.	0.		0.	0.	0.	0.
102	0.	0.	0.	0.	0.		0.	0.	0.	0.
114	0.	0.	0.	0.	0.		0.	0.	0.	0.

TABLE X (continued)

[illegible]

	AVSTAND(KM)	275	285	295	VINKEL(GRADER)	315	325	335	345	355
3	0.	0.	0.	0.	0.	0	2.	4.	6.	8.
9	0.	0.	0.	0.	0.	0	0.	0.	0.	3.
15	0.	0.	0.	0.	0.	0	0.	0.	18.	15.
21	0.	0.	0.	0.	0.	0	0.	2.	138.	69.
27	0.	0.	0.	0.	0.	0	0.	14.	125.	164.
33	0.	0.	0.	0.	0.	0	0.	0.	80.	104.
39	0.	0.	0.	0.	0.	0	0.	6.	137.	290.
45	0.	0.	0.	0.	0.	0	0.	41.	232.	284.
51	0.	0.	0.	0.	0.	0	0.	93.	318.	291.
57	0.	0.	0.	0.	0.	0	0.	52.	305.	41.
66	0.	0.	0.	0.	0.	0	0.	48.	95.	1038.
78	0.	0.	0.	0.	0.	0	0.	0.	346.	2000.
90	0.	0.	0.	0.	0.	0	0.	0.	0.	0.
102	0.	0.	0.	0.	0.	0	0.	0.	0.	147.
114	0.	0.	0.	0.	0.	0	0.	0.	0.	1060.

contain all of the iodine that is deposited on to it. The experimentally determined half-life of ^{131}I on the ground, 5.5 days, mentioned earlier, confirms this assumption. For conservative reasons, however, the intake is considered equal to that of grazing cows.

The intake of ^{131}I via inhalation occurs from direct inhalation from the plume and/or from ground air contaminated during deposition. This is, in contrast to intake from forage, a pulsed injection of ^{131}I into blood via the lung.

The total intake via inhalation follows the same distance dependence as the area content given in figures 2 and 3. Outdoors the total intake is about 0.5% of the total uptake during the first day from grazing. (FOA, 1980). Data are very limited on the reduction of air activity concentration indoors, but tends to be dependent on the particle size. According to data given by FOA (1980) and SSI (1980), the concentration values indoors for small particles ($<2\mu\text{m}$) are 80-100% of those outdoors. For greater particles ($2-10\mu\text{m}$) the values is between 90% and 20%. For particles greater than $5-10\mu\text{m}$ the figures is less than 20%. In this study the figure has been set to 60%.

This fraction, therefore inhales 60% of those grazing in the fields.

- 2) For the 0.15 fraction the forage intake is the same as for the 0.1 fraction. The amount of ^{131}I inhaled is greater due to the fact that these cows are outdoors during daytime.
- 3) For the 0.75 fraction the daily intake from grazing is the same as that for the other fractions. The intake from inhalation is maximal both during day and night.
- 4) During the "Winter" period all cows are inside cow-houses. Intake of ^{131}I thus entirely comes from inhalation, since the food supplied has comparatively low ^{131}I -contamination.
- d) The area (m^2) that is grazed per day differ much from various sources in the literature. A mean value, which is also found in the study of Booth et al. is $67\text{ m}^2/\text{day}$. This is thus used in the calculations and is called Y. The breathing rate for cows is approximately 55 l/min. This gives rise to a single intake of, as mentioned above, 0.5% of the activity intake during the first day from grazing (67 m^2).

- e) Not all of the milk produced is delivered to the dairies. Some is lost during the handling and transport, and some is consumed on the site of production, that is at the farm, which is an important factor for the highest individual doses. The fraction that is delivered is 0.91 and is called f_d .
- f) All of the delivered milk is not used for milk production. Some is used for making cheese and some is used for other purposes, for example production of dried milk. The fraction that is used for production of consumption milk (and cream) is in Sweden around 0.5, and is called f_p .
- g) The excretion into milk following intake by grazing of the contaminated pasture can be expressed using the function $\epsilon_m(t)$ described by equation (3). The activity input rate follows the decay of ^{131}I and the initial input rate is given by figures 2 and 3. For the inhalation case, $\epsilon_m(t)$ is obtained from the calculation of an single pulse injection of unit activity rate into the blood 1 compartment, whereafter the expression (3) applies.

The dose calculations can now be carried out. These are divided into two types:

- 1) Collective doses
- 2) Individual doses

For the collective dose case, the variation of ^{131}I levels per liter milk from cow to cow has been omitted due to the large cow population which in this area is approximately 94 000 individuals. The uncertainty in absorbed dose figures are assumed mainly to be due to the variation in time between production and consumption. In the individual dose case is, again for conservative reasons, only consumption on the site of production considered. In this case no delay between production and consumption is assumed, and the variations in dose figures are explained by variations between individual cows.

The dose figures apply to the intake of ^{131}I from milk produced on day number $\underline{t}$ from the time of the reactor accident.

For the collective dose case the activity available for intake produced in sector i on day number t is, in the "summer" case for the fraction

a) Out grazing (fraction 0.75)

$$A_i^1(t) = A_{g,i}(0) \cdot 0.75 \cdot N_i \cdot Y \cdot f_d \cdot f_p \cdot (\epsilon_m(t) + 0.005 \cdot \epsilon_{m, \text{lung}}(t)) \cdot e^{-\lambda T} \quad (9)$$

where ϵ_m and $\epsilon_{m, \text{lung}}$ corresponds to intake via forage and inhalation, respectively. T is the time between production and consumption and is, in southern Sweden, for milk and cream 5±2 days.

b) Fed outside the cow-house (fraction 0.15)

$$A_i^2(t) = A_{g,i}(0) \cdot 0.15 \cdot N_i \cdot Y \cdot f_d \cdot f_p \cdot (\epsilon_m(t) + 0.8 \cdot 0.005 \cdot \epsilon_{m, \text{lung}}(t)) \cdot e^{-\lambda T} \quad (10)$$

where the reduction of inhalation to 0.8 comes from the fact that the cows partly are inside the cow-house.

c) Cows that never are outside the cow-house (fraction 0.10)

$$A_i^3(t) = A_{g,i}(0) \cdot 0.10 \cdot N_i \cdot Y \cdot f_d \cdot f_p \cdot (\epsilon_m(t) + 0.6 \cdot 0.005 \cdot \epsilon_{m, \text{lung}}(t)) \cdot e^{-\lambda T} \quad (11)$$

The factor 0.6 for inhalation follows from the fact that these cows are never outdoors.

The total activity available is, of course

$$A_i(t) = A_i^1(t) + A_i^2(t) + A_i^3(t) \quad (12)$$

For the "winter" case the following expression applies (fraction 1.0 indoors)

$$A_i(t) = A_{g,i}(0) \cdot N_i \cdot Y \cdot f_d \cdot f_p \cdot 0.005 \cdot \epsilon_{m, \text{lung}}(t) \cdot e^{-\lambda T} \quad (13)$$

The total collective dose can now be calculated. For the intake of ^{131}I inside the whole 120 km (radius) area the collective dose arising from the intake of milk that is produced during t days is found by multiplying equation (12) (summer) or (13) winter by the sum of equations (8),

The expressions for A_{is} , A_{ib} and A_{iv} (as functions of A_i) are found in equation (6).

We then conclude:

The collective dose to infants for organ (o)

$$CD_{s,o} = \sum_{it} \frac{A_i(t) \cdot f_s \cdot F_s \cdot C_{s,o}}{f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v} = \frac{f_s \cdot F_s \cdot C_{s,o}}{f_s \cdot F_s + f_b \cdot F_b + f_v \cdot F_v} \cdot \sum_{it} A_i(t) \quad (14)$$

where $A_i(t)$ is given by equations (12) or (13)

The collective doses for children and adults are found in the same way.

The factor preceding the double sum sign can be calculated using the values given in table IX. The final expression for the collective doses will be: (activity in Curies). Note: contribution from Denmark is, due to lack of data, not considered.

A) Absorbed dose to thyroid:

$$\text{For infants} \quad CD_{s,th} = 13000 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (15)$$

$$\text{For children} \quad CD_{b,th} = 11000 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (16)$$

$$\text{For adults} \quad CD_{v,th} = 14000 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (17)$$

$$\text{Sum} \quad CD_{th} = 38000 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (18)$$

The corresponding figures for weighted whole body doses are:

$$\text{For infants} \quad CD_{s,w} = 400 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (19)$$

$$\text{For children} \quad CD_{b,w} = 340 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (20)$$

$$\text{For adults} \quad CD_{v,w} = 420 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (21)$$

$$\text{Sum} \quad CD_w = 1160 \cdot \sum_{it} A_i(t) \quad (\text{man Sv}) \quad (22)$$

For the individual doses the situation is different. The following assumption are made:

- a) The milk is consumed on the site of production without delay.
 - b) No mixing of milk between cows is made. This will require that individual variations in iodine metabolism between cows is considered, and that the type of cow that gives the highest excretion of ^{131}I into milk is considered as source for the radionuclide. The functions $\epsilon_m(t)$ and $\epsilon_{m,\text{lung}}(t)$ then has to be multiplied with $11/6 \approx 1.8$ to be applicable in this case.
- The maximum thyroid dose will appear in infants and is calculated as (activity in Curies).

$$D_{\text{max},s}^{\text{th}} = A_{\text{g,max}}(0) \cdot Y \cdot 1.8 (\epsilon_m(t) + 0.005 \cdot \epsilon_{m,\text{lung}}(t)) \cdot \frac{0.5}{16} \cdot C_{s,\text{th}} \quad (23)$$

or, with the values substituted

$$D_{\text{max},s}^{\text{th}} = A_{\text{g,max}}(0) \cdot (\epsilon_m(t) + 0.005 \cdot \epsilon_{m,\text{lung}}(t)) \cdot 87000 \text{ (Sv)} \quad (24)$$

This equation is then integrated for the number of days that is to be included.

In this expression, no consideration is given to individual variations in the area grazed by the cow and the individual variations in thyroid uptake and retention in the child. Such variations would naturally affect the individual dose figures. These can, however, easily be corrected by simply multiplying the figures with the appropriate factors.

The maximum weighted whole-body dose is also found in the infant, and a similar analysis will result in

$$D_{\text{max},s}^{\text{w}} = A_{\text{g,max}}(0) \cdot (\epsilon_m(t) + 0.005 \cdot \epsilon_{m,\text{lung}}(t)) \cdot 2600 \text{ (Sv)} \quad (25)$$

The results are, for each of the eight cases described previously (summer, winter, Pasquill D-F, wet and dry deposition) calculated for variation of the wind direction with angle θ from 5, 15, 25...to 355 degrees. The results are then given in figures 9-16 as collective

Figure 9

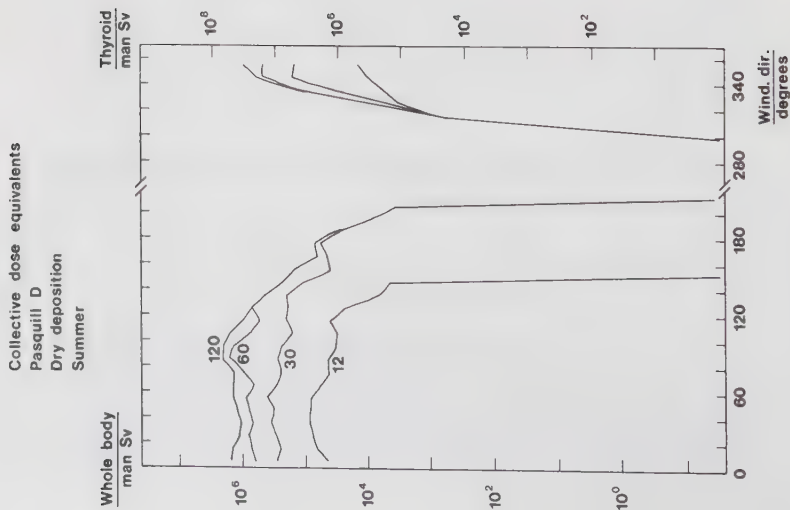


Figure 10

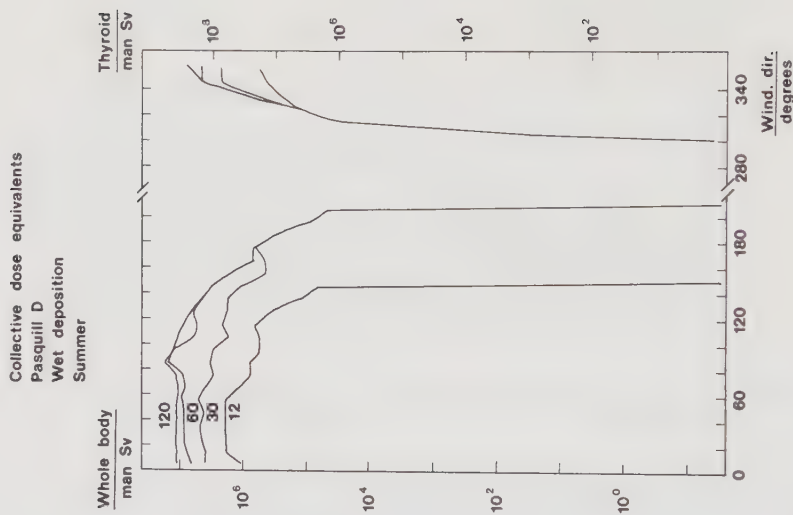


Figure 11

Collective dose equivalents

Pasquill F

Dry deposition

Summer

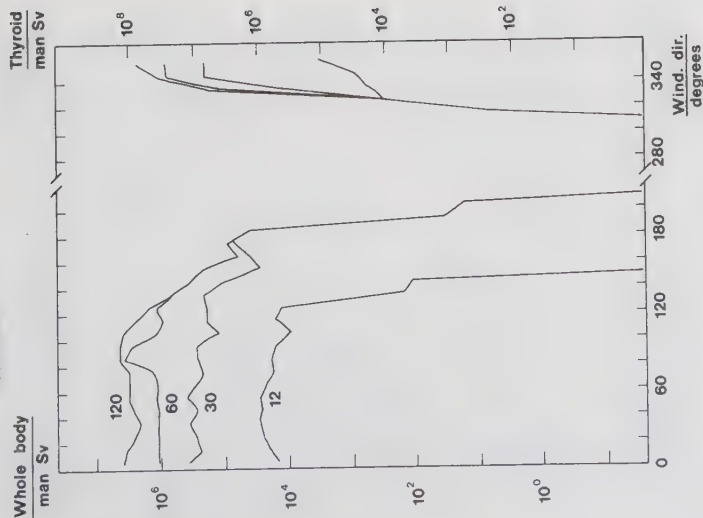


Figure 12

Collective dose equivalents

Pasquill F

Wet deposition

Summer

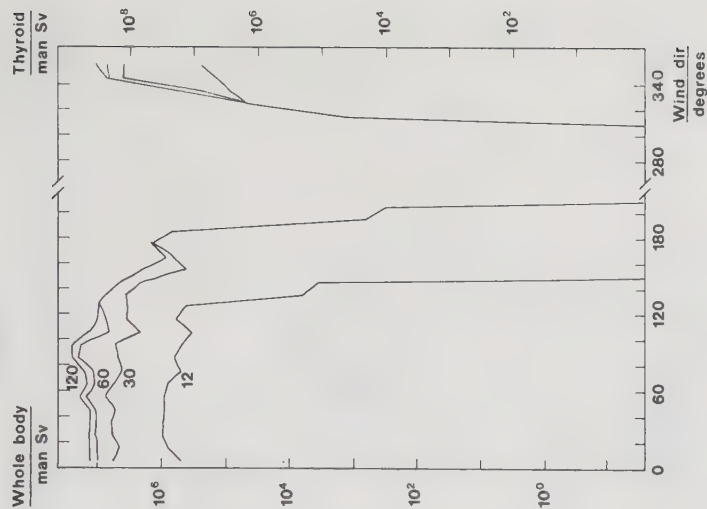


Figure 13

Collective dose equivalents

Pasquill D

Dry deposition

Winter

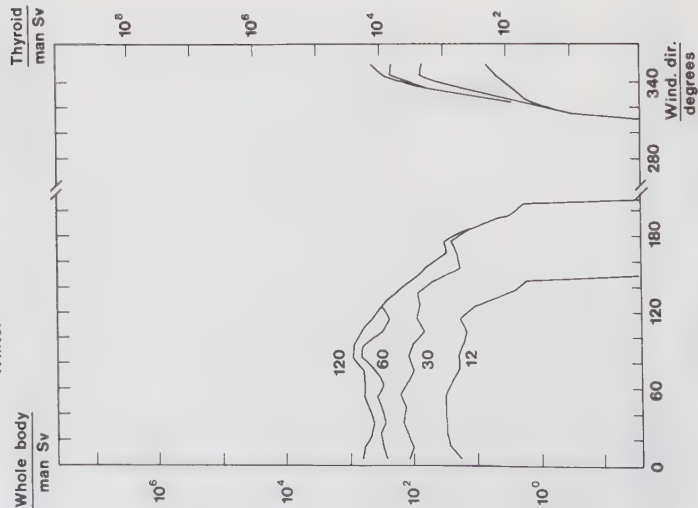


Figure 14

Collective dose equivalents

Pasquill D

Wet deposition

Winter

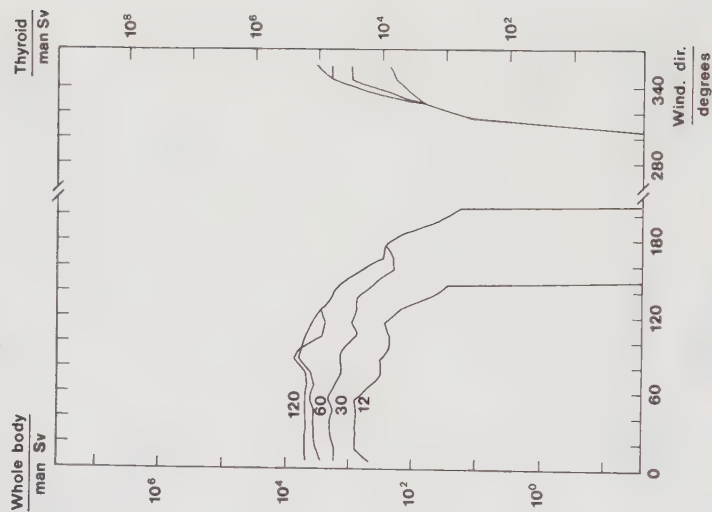


Figure 15

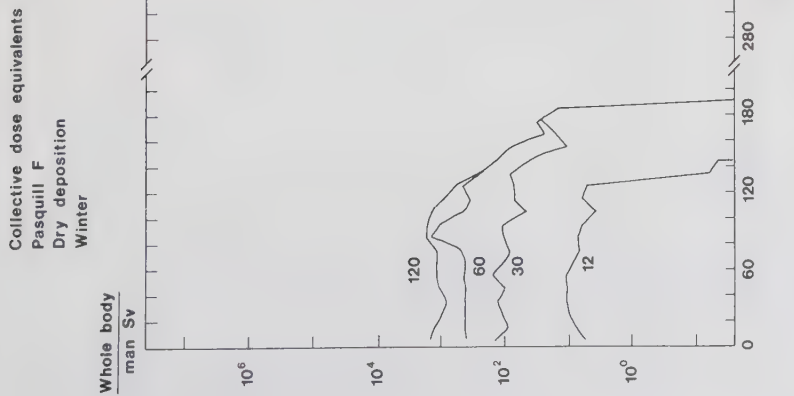
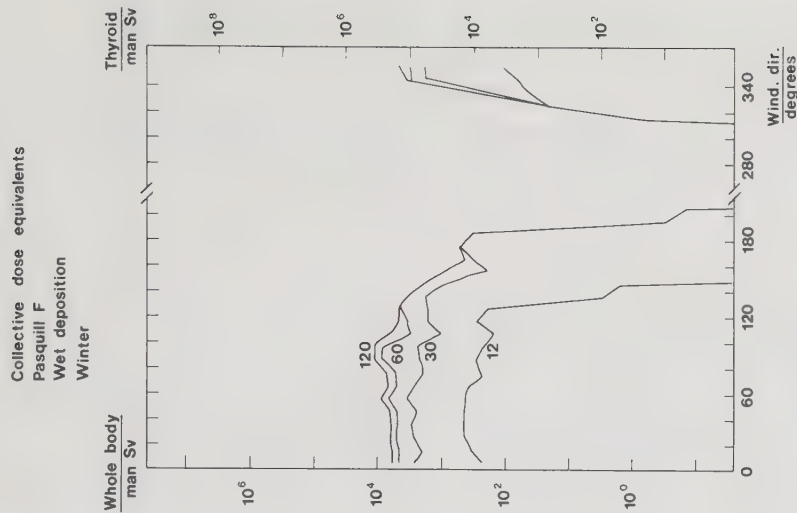


Figure 16



dose equivalent as a function of wind direction. The maximum individual dose does not vary with wind direction, unless no cows are present in the sectors included. These figures are given in table XI below.

The individual curves in figures 9-16 represents collective dose equivalents arising from the intake of milk produced within 12, 30, 60 and 120 km from the Barsebäck nuclear power plant.

Table XI

a) Maximum individual thyroid dose equivalent (Sv)

	Summer		Winter	
	Dry dep.	Wet dep.	Dry dep.	Wet dep.
Pasquill D	4490	179600	1.1	45
Pasquill F	3050	85300	0.8	21

b) Maximum individual weighted whole body dose equivalent (Sv)

	Summer		Winter	
	Dry dep.	Wet dep.	Dry dep.	Wet dep.
Pasquill D	135	5390	0.03	1.3
Pasquill F	92	2560	0.02	0.6

c) Maximum individual whole body dose equivalent (ICRP 2) (Sv)

	Summer		Winter	
	Dry dep.	Wet dep.	Dry dep.	Wet dep.
Pasquill D	2	77	0.0004	0.02
Pasquill F	1.3	37	0.0003	0.01

It should be stressed that the figures given in the diagrammes 9-16 and table XI are based on consumption of milk every day during 80 days following the accident. These values are very close to those obtained when integration of equations 15-25 to infinity is made. (Eighty days is almost ten half-lives of ^{131}I .)

All dose figures will, of course, be reduced if the milk production is stopped at an earlier time. In table XII reduction factors for all dose figures are given. These figures express the reduction in dose

(equivalents) that are obtained if the consumption of milk produced within the contaminated area is stopped at the times given in the table. These reduction factors differ from summer to winter due to the different pattern of input of ^{131}I into the dairy cow. They are obtained from integration of the quantity ϵ_m and $\epsilon_{m,\text{lung}}$ given in equation (3). From this table, it is also possible to calculate the "complementary" reduction factor,

$$1 - \int_0^t \epsilon_m(t) dt = \int_t^\infty \epsilon_m(t) dt$$

which will express the reduction in collective doses when the milk production and consumption is started again after t days after the accident. It is also possible to calculate reduction factors with regard to a limited time interval during which milk is produced and consumed, that is

$$\int_{t_1}^{t_2} \epsilon_m(t) dt$$

Table XII

Time (days)	Reduction factors	
	Summer	Winter
1	0.018	0.21
2	0.065	0.59
4	0.19	0.90
8	0.42	0.97
16	0.70	0.99
32	0.92	0.99
64	0.99	1.00
80	1.00	1.00

All values in figures 9-16 refer to collective dose equivalents for the whole population. From equations 15-22 one could then calculate reduction factors for the specific populations of infants, children and adults. For convenience, however, these are in table XIII given as factors relative to the whole population.

Table XIII

Reduction factors for collective doses

Age group	Reduction factor
Infants < 5 years	0.34
Children 5-15 years	0.29
Adults > 15 years	0.37

The maximum individual dose equivalents given in table XI a,b all refer to infants (<5 years) due to the great thyroid dose conversion factor for this group. The corresponding figures for children and adults are, of course, lower and can be calculated from table IX. The reduction factors are summarized in table XIV.

Table XIV

Reduction factors for individual doses

Age group	Reduction factors
Children 5-15 years	0.39
Adults > 15 years	0.09

It should be stressed that these figures are not applicable in the ICRP 2 case (table XI c).

Consequences of the BWR 1 accident.

As can be seen from the results, the dominating dose figures are those for the thyroid. The dominating radiological effect is accordingly radiation effects on thyroid function and induction of cancer in the thyroid gland.

In the case of maximum individual doses, the irradiation of the thyroid from sources other than from ^{131}I via milk by far is the most important factor. For instance, during winter-time, the direct inhalation of ^{131}I by humans will result in higher doses to the thyroid than the consumption of milk. As the purpose of this work only is to estimate the consequences from the intake via milk, this analysis is omitted.

For the collective doses, however, the situation is different. A large number of people will be irradiated only due to milk intake, because of the distribution of milk from the dairies.

Thyroid cancer induced by ionizing radiation has been studied in these categories:

- a) A-bomb survivors
- b) Inhabitants from the Marshall islands, which were subjected to fallout from nuclear weapon tests.
- c) Patients exposed to radiation given externally or internally.

Results from these studies are found in the BEIR-report (BEIR,1972) and in the UNSCEAR report, 1977.

The excess rate is found to be in the order of $(50-150) \cdot 10^{-4} \cdot \text{Sv}^{-1}$. Only a small fraction of the induced cancers is fatal; figures ranging from 3% to 10% can be found in the literature. There are vague tendencies that the excess rates are higher when the individuals irradiated are young, but the differences are too small to influence the results for a large population.

There is, however, clear evidence that (Strand, 1978) the latency time between irradiation and disease is dependent on the age of the irradiated individual and can be expressed as:

$$\text{Latency time} = 9.4 + 0.27 \cdot \text{Age (years)}$$

This age-dependent latency time will decrease the number of cancers that actually will be discovered to about half, as the whole population is irradiated.

Approximately 10^6 persons are living within the 120 km zone. Calculation of effects using the data from this work would result in the maximum and minimum number of induced thyroid cancers given below. The assumption for this analysis is naturally that the contaminated milk is equally distributed within the zone. These figures are also based on consumption during 80 days. For other periods of time, table XII should be used to reduce the figures.

For a 100% iodine release in the summer period, the maximum collective dose equivalent is in the order of $10^8 - 10^9$ man Sv. Assuming that this collective dose is equally distributed within the irradiated population, it would probably result in a limited number of thyroid cancer cases. This follows from the fact that the individual doses in this case are high enough ($10^2 - 10^3$ Gy) to kill all living thyroid cells, thus resulting in nonfunctioning thyroids rather than cancer.

The actual situation would probably consist of a nonuniformly distributed collective dose equivalent with regard both to the population and age distribution. In order to estimate the "expected" excess cancer rate, the knowledge of this dose distribution, which primarily is dependent on the distribution of milk between and from the dairies, is required. (At present, this information is being prepared for insertion into a geographical model. Results will be reported in a later part of this article series).

In winter time, the dose values are considerably lower, and application of the excess rate figures from BEIR and UNSCEAR result in 500 - 1500 cases of thyroid cancer.

It should once again be stressed that only a minor fraction of these cancers will be fatal.

These figures should be compared to those obtained when using the "risk estimate" factors published in a recent thesis by Holm, (Holm, 1980) which deals with patients treated with ^{131}I for different malfunctions of their thyroid. Roughly the figures from his thesis result in risk factors of about a factor 100 lower than those from BEIR and UNSCEAR. The number of thyroid cancers should thus be reduced to 0.01 of those given above if Holm's figures are used in the risk analysis.

Another important type of cancer that should be considered is leukaemia. Figures from BEIR and UNSCEAR indicate an excess rate of $(15 - 25) \cdot 10^{-4} \cdot \text{Sv}^{-1}$. The mean latency time is around ten years. For these calculations, the dose conversion factor that should be used is that for bone-marrow, which can be found in table VIII (SFR, 1979).

The most probable weather situation during summer would give a maximum number of leukaemia cases of about 300. The absolute maximum value would occur for the improbable weather situation of class Pasquill F together with wet deposition during summer and would be

about 4000 cases. During winter-time, the collective dose equivalent is too low to influence the leukaemia incidence.

Uncertainties in the results.

Prior to the uncertainty discussion, the most basic assumptions and approximations made must once again be repeated in order to simplify the interpretation of the results. These are:

- 1) 100% of the iodine in the core is released (46 MCi).
- 2) The activity is released as a pulse of a duration that is short compared to the physical half-life of ^{131}I (8.05 d)
- 3) The wind direction and speed is assumed to be constant during the ground deposition time.
- 4) The deposition is assumed to be either dry or wet; no "hot or cold-spots" are considered
- 5) All activity/m² that is deposited on the ground is available for intake by the cow
- 6) The percentage of the total population for infants, children and adults, respectively, follows that for the national Swedish statistics.

The main factor giving rise to uncertainty in the collective dose equivalent values is the variation of time between production and consumption of milk. The word "production" in this case refers to the production of milk in the udder of the cow. The mean time from production to consumption is five days during "normal" weeks of the year. The shortest time possible is around two days and the longest about eight days. The " ± 1 S.D" limits are therefore set to 2 days. A small fraction of the population therefore receive doses that are somewhat lower. On the other hand, this is compensated by the other fraction that receives higher doses. The conclusion must be that it is not very probable that, considering the basic assumption, the uncertainty in the collective dose equivalent figures is more than 20%.

In the individual dose equivalent cases, uncertainties in many parameters could affect the result. Variations in the metabolism of iodine both in the cow and in humans can naturally affect the result, as well as variation in the area grazed by the individual cow. An important factor that up to now not has been considered is the amount of

¹³¹I that is administered to the cow by its drinking-water. Iodine is very soluble in water and is sooner or later dissolved in vast volumes of ground water, which is in contact with wells or other sources for drinking-water. There is a possibility that the drinking-water intake is of importance in the cases when a local water-supply is located at the farm. The well that could function as a source for ¹³¹I intake into the cow, must, however, be shallow enough so that a rapid transport of the radionuclide from the ground surface into the well is obtained. On the other hand, it must be deep enough to be sufficient for the daily water consumption for cows, which is in the order of 50 - 60 liters/day. This situation is then probable only for a farm with a very small stock of cattle, which in these areas is seldom found.

It is almost impossible to give figures representing some kind of error in the individual dose figures, since the uncertainties in the input parameters are strongly connected with variations between individual organisms and geographic sites of contamination. Fortunately, all corrections of dose figures in this case are linear with the variation of the input parameters and could easily be applied to a specific situation if the input data is accurately known.

This completes part III: Application on ¹³¹I in the food-chain grass-cow-milk-man following a BWR 1 accident.

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